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- (54) Title: 3-HYDROXY-4-ARYL-5-PYRAZOLINE DERIVATIVES AS HERBICIDES
- (57) Abstract

Compounds of formula (I), in which the substituents are as defined in claim 1, are suitable for use as herbicides, in particular in combination with herbicide-antagonistically effective compounds.

$$\begin{array}{c|c}
R_5 & R_1 \\
R_4 & R_2
\end{array}$$

$$\begin{array}{c|c}
R_2 & (I)
\end{array}$$

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3-HYDROXY-4-ARYL-5-PYRAZOLINE DERIVATIVES AS HERBICIDES

The present invention relates to novel herbicidally active 3-hydroxy-4-aryl-5-oxopyrazoline derivatives, to processes for their preparation, to compositions which comprise these compounds and may additionally comprise safeners, and to the use of these compounds as herbicides for controlling weeds and grasses, in particular in crops of useful plants.

3-Hydroxy-4-aryl-5-oxopyrazoline derivates having herbicidal action are described, for example, in EP-A-0 508 126, WO 96/25395 and WO 96/21652. We have now found novel 3-hydroxy-4-aryl-5-oxopyrazoline derivatives having herbicidal properties.

The present invention thus provides compounds of the formula I

$$R_{4}$$
 N_{1}
 R_{4}
 R_{3}
 R_{4}
 R_{3}
 R_{4}
 R_{3}
 R_{4}
 R_{4}
 R_{4}
 R_{4}
 R_{5}
 R_{4}
 R_{5}
 R_{2}
 R_{3}
 R_{4}
 R_{4}
 R_{5}
 R_{5

in which

 R_1 , R_2 and R_3 independently of one another are halogen, nitro, cyano, C_1 - C_4 alkyl, C_2 - C_4 alkenyl, C_2 - C_4 alkynyl, C_1 - C_4 haloalkyl, C_2 - C_6 haloalkenyl, C_3 - C_6 cycloalkyl, halogen-substituted C_3 - C_6 cycloalkyl, C_1 - C_6 alkoxyalkyl, C_1 - C_6 alkylthioalkyl, hydroxyl, mercapto, C_1 - C_6 alkoxy, C_3 - C_6 alkenyloxy, C_3 - C_6 alkynyloxy, C_1 - C_4 alkylcarbonyl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfinyl, amino, C_1 - C_4 alkylamino or di(C_1 - C_4 -alkyl)amino;

R₄ and R₅ together are a group

$$-C-R_{6}(R_{7})-O-C-R_{8}(R_{9})-C-R_{10}(R_{11})-C-R_{12}(R_{13})- \\ \hspace{2cm} (Z_{1}),$$

$$-C-R_{14}(R_{15})-C-R_{16}(R_{17})-O-C-R_{18}(R_{19})-C-R_{20}(R_{21})-$$
 (Z₂), or

$$-C-R_{22}(R_{23})-C-R_{24}(R_{25})-C-R_{26}(R_{27})-O-C-R_{28}(R_{29})-$$
 (Z₃);

in which R_6 , R_7 , R_8 , R_9 , R_{10} , R_{11} , R_{12} , R_{13} , R_{14} , R_{15} , R_{16} , R_{17} , R_{18} , R_{19} , R_{20} , R_{21} , R_{22} , R_{23} , R_{24} , R_{25} , R_{26} , R_{27} , R_{28} , and R_{29} independently of one another are hydrogen, halogen, C_1 - C_4 alkyl

or C₁-C₄haloalkyl where an alkylene ring, which together with the carbon atoms of the groups Z₁, Z₂ or Z₃ contains 2 to 6 carbon atoms and may be interrupted by oxygen, may either be fused or spiro-linked to the carbon atoms of the groups Z₁, Z₂ or Z₃, or where this alkylene ring bridges at least one ring atom of the groups Z_1 , Z_2 or Z_3 ; G is hydrogen, $-C(X_1)-R_{30}$, $-C(X_2)-X_3-R_{31}$, $-C(X_4)-N(R_{32})-R_{33}$, $-SO_2-R_{34}$, an alkali metal, alkaline earth metal, sulfonium or ammonium cation or -P(X₅)(R₃₅)-R₃₆; $X_1,\,X_2,\,X_3,\,X_4$ and X_5 independently of one another are oxygen or sulfur; and R_{30} , R_{31} , R_{32} , R_{33} , R_{34} , R_{35} and R_{36} independently of one another are hydrogen, C_1 - C_5 alkyl, C_1 - C_5 haloalkyl, C_2 - C_5 alkenyl, C_1 - C_5 alkoxyalkyl, C_3 - C_6 cycloalkyl or phenyl, and R₃₄ is additionally C₂-C₂₀alkenyl, C₂-C₂₀alkenyl substituted by halogen, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, alkoxy, thioalkyl, alkylthiocarbonyl, alkylcarbonylthio. alkylsulfonyl, alkylsulfoxyl, alkylaminosulfonyl, dialkylaminosulfonyl, alkylsulfonyloxy, alkylsulfonylamino, alkylamino, dialkylamino, alkylcarbonylamino, dialkylcarbonylamino, alkyl-alkylcarbonylamino, cyano, (C3-C7)cycloalkyl, (C3-C7)heterocyclyl, trialkylsilyl, trialkylsilyloxy, phenyl, substituted phenyl, heteroaryl or substituted heteroaryl, C2-C20alkynyl, C2-C20alkynyl substituted by halogen, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, alkoxy, thioalkyl, alkylthiocarbonyl, alkylcarbonylthio, alkylsulfonyl, alkylsulfoxyl, alkylaminosulfonyl, dialkylaminosulfonyl, alkylsulfonyloxy, alkylsulfonylamino, alkylamino, dialkylamino, alkylcarbonylamino, dialkylcarbonylamino, alkylalkylcarbonylamino, cyano, (C₃-C₇)cycloalkyl, (C₃-C₇)heterocyclyl, trialkylsilyloxy, phenyl, substituted phenyl, heteroaryl or substituted heteroaryl, (C1-C7)cycloalkyl, (C1-C₇)cycloalkyl substituted by halogen, haloalkyl, (C₁-C₆)alkyl, alkoxy, alkylcarbonyloxy, thioalkyl, alkylcarbonylthio, alkylamino, alkylcarbonylamino, trialkylsilyl or trialkylsilyloxy, heteroaryl, heteroaryl substituted by halogen, haloalkyl, nitro, cyano, (C1-C6)alkyl, alkoxy, alkylcarbonyloxy, thioalkyl, alkylcarbonylthio, alkylamino, alkylcarbonylamino, trialkylsilyl or trialkylsilyloxy, heteroaryloxy, substituted heteroaryloxy, heteroarylthio, substituted heteroarylthio, heteroarylamino, substituted heteroarylamino, diheteroarylamino, substituted diheteroarylamino, phenylamino, substituted phenylamino, diphenylamino, substituted diphenylamino, cycloalkylamino, substituted cycloalkylamino, dicycloalkylamino, substituted dicycloalkylamino, cycloalkoxy or substituted cycloalkoxy, and salts and diastereomers of the compounds of the formula I.

In the above definitions, halogen is to be understood as meaning fluorine, chlorine, bromine and iodine, preferably fluorine, chlorine and bromine. The alkyl groups in the definitions of the substituents are, for example, methyl, ethyl, n-propyl, isopropyl, n-butyl, sec-butyl, isobutyl or tert-butyl, and the isomeric pentyls and hexyls. Suitable cycloalkyl substituents

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contain 3 to 6 carbon atoms and are, for example, cyclopropyl, cyclobutyl, cyclopentyl or cvclohexyl. These may be mono- or polysubstituted by halogen, preferably by fluorine, chlorine or bromine. Alkenyl is to be understood as meaning, for example, vinyl, allyl, methallyl, 1-methylvinyl or but-2-en-1-yl. Alkynyl is, for example, ethinyl, propargyl, but-2-in-1-yl, 2-methylbutin-2-yl or but-3-in-2-yl. Haloalkyl groups preferably have a chain length of 1 to 4 carbon atoms. Haloalkyl is, for example, fluoromethyl, difluoromethyl, trifluoromethyl, chloromethyl, dichloromethyl, trichloromethyl, 2,2,2-trifluoroethyl, 2-fluoroethyl, 2chloroethyl, pentafluoroethyl, 1,1-difluoro-2,2,2-trichloroethyl, 2,2,3,3-tetrafluoroethyl and 2,2,2-trichloroethyl; preferably trichloromethyl, difluorochloromethyl, difluoromethyl, trifluoromethyl and dichlorofluoromethyl. Suitable haloalkenyls are alkenyl groups which are mono- or polysubstituted by halogen, halogen being fluorine, chlorine, bromine and iodine and in particular fluorine and chlorine, for example 2,2-difluoro-1-methylvinyl, 3fluoropropenyl, 3-chloropropenyl, 2,3,3-trifluoropropenyl, 2,3,3trichloropropenyl and 4,4,4-trifluorobut-2-en-1-yl. Among the C2-C6alkenyl groups which are mono-, di- or trisubstituted by halogen, preference is given to those having a chain length of 3 to 5 carbon atoms. Alkoxy groups preferably have a chain length of 1 to 6 carbon atoms. Alkoxy is, for example, methoxy, ethoxy, propoxy, isopropoxy, n-butoxy, isobutoxy, secbutoxy and tert-butoxy, and the isomeric pentyloxy and hexyloxy radicals; preferably methoxy and ethoxy. Alkylcarbonyl is preferably acetyl or propionyl. Alkoxycarbonyl is, for example, methoxycarbonyl, ethoxycarbonyl, propoxycarbonyl, isopropoxycarbonyl, nbutoxycarbonyl, isobutoxycarbonyl, sec-butoxycarbonyl or tert-butoxycarbonyl; preferably methoxycarbonyl or ethoxycarbonyl. Alkylthio groups preferably have a chain length of 1 to 4 carbon atoms. Alkylthio is, for example, methylthio, ethylthio, propylthio, isopropylthio, nbutylthio, isobutylthio, sec-butylthio or tert-butylthio, preferably methylthio and ethylthio. Alkylsulfinyl is, for example, methylsulfinyl, ethylsulfinyl, propylsulfinyl, isopropylsulfinyl, nbutylsulfinyl, isobutylsulfinyl, sec-butylsulfinyl, tert-butylsulfinyl; preferably methylsulfinyl and ethylsulfinyl. Alkylsulfonyl is, for example, methylsulfonyl, ethylsulfonyl, propylsulfonyl, isopropylsulfonyl, n-butylsulfonyl, isobutylsulfonyl, sec-butylsulfonyl or tert-butylsulfonyl; preferably methylsulfonyl or ethylsulfonyl. Alkylamino is, for example, methylamino, ethylamino, n-propylamino, isopropylamino or the isomeric butylamines. Dialkylamino is, for example, dimethylamino, methylethylamino, diethylamino, n-propylmethylamino, dibutylamino and diisopropylamino. Alkoxyalkyl groups preferably have 1 to 6 carbon atoms. Alkoxyalkyl is, for example, methoxymethyl, methoxyethyl, ethoxymethyl, ethoxyethyl, n-propoxymethyl, n-propoxyethyl, isopropoxymethyl or isopropoxyethyl. Alkylthioalkyl is, for example, methylthiomethyl, methylthioethyl, ethylthiomethyl, ethylthioethyl, n-propylthiomethyl, n-propylthioethyl, isopropylthiomethyl, isopropylthioethyl,

butylthiomethyl, butylthioethyl or butylthiobutyl. Phenyl may be substituted. In this case, the substituents may be in the ortho, meta and/or para position. The substituents are preferably located in the positions ortho and para to the site where the ring is attached.

The halogen, alkyl, cycloalkyl, alkoxy, alkylthio, alkylcarbonyl, alkylsulfonyl and (di)alkylamino radicals which may be present in the radicals R₃₄, in particular -SO₂R₃₄ (G), are derived from the corresponding radicals mentioned above. Preferred heterocyclyl radicals are those containing 1 or 2 heteroatoms, for example N, S or O. They are usually saturated. Heteroaryl radicals are customarily aromatic heterocycles which preferably contain 1 to 3 heteroatoms, such as N, S and O. Examples of suitable heterocycles and heteroaromatics are: pyrrolidine, piperidine, pyran, dioxane, azetidine, oxetan, pyridine, pyrimidine, triazine, thiazole, thiadiazole, imidazole, oxazole, isoxazole and pyrazine, furan, morpholine, piperazine, pyrazole, benzoxazole, benzothiazole, quinoxaline and quinoline. These heterocycles and heteroaromatics may also be substituted, for example by halogen, alkyl, alkoxy, haloalkyl, haloalkoxy, nitro, cyano, thioalkyl, alkylamino or phenyl. The C₂-C₂₀alkenyl and alkynyl groups R₃₄ may be mono- or polyunsaturated. They preferably contain 2 to 12, in particular 2 to 6, carbon atoms. For illustration, suitable groups -SO₂R₃₄ are given in the example below:

Alkali metal cations, alkaline earth metal cations or ammonium cations for the substituent G are, for example, the cations of sodium, potassium, magnesium, calcium and ammonium. Preferred sulfonium cations are, in particular, trialkylsulfonium cations, where the alkyl radicals each preferably contain 1 to 4 carbon atoms.

The free valency on the left hand of the groups Z_1 , Z_2 and Z_3 is linked to the 1-position and the free valency on the right hand is linked to the 2-position of the pyrazoline ring.

Compounds of the formula I, in which an alkylene ring which, together with the carbon atoms of the groups Z_1 , Z_2 and Z_3 contains 2 to 6 carbon atoms may be fused or spirolinked to the groups Z_1 , Z_2 and Z_3 , have, for example, the following structure:

$$R_2$$
 (spiro-linked) or R_3 (fused).

Compounds of the formula I, in which in the groups Z_1 , Z_2 or Z_3 an alkylene ring bridges at least one ring atom of the groups Z_1 , Z_2 or Z_3 , have, for example, the following structure:

R₄ and R₅ together are in particular a group

$$-C-R_6(R_7)-O-C-R_8(R_9)-C-R_{10}(R_{11})-C-R_{12}(R_{13})-$$
 (Z₁),

$$-C-R_{22}(R_{23})-C-R_{24}(R_{25})-C-R_{26}(R_{27})-O-C-R_{28}(R_{29})-$$
 (Z₃);

in which R_6 , R_7 , R_8 , R_9 , R_{10} , R_{11} , R_{12} , R_{13} , R_{14} , R_{15} , R_{16} , R_{17} , R_{18} , R_{19} , R_{20} , R_{21} , R_{22} , R_{23} , R_{24} , R_{25} , R_{26} , R_{27} , R_{28} and R_{29} independently of one another are hydrogen, halogen, C_1 - C_4 alkyl or C_1 - C_4 haloalkyl, where an alkylene ring which, together with the carbon atoms of the groups

 Z_1 , Z_2 and Z_3 , contains 3 to 6 carbon atoms may be fused or spiro-linked to the groups Z_1 , Z_2 and Z_3 .

Among the compounds of the formula I, preference is given to those in which G is hydrogen. In a particularly preferred group of compounds of the formula I, R_4 and R_5 together form a group Z_2 . Also of particular interest are compounds of the formula I in which R_1 , R_2 and R_3 independently of one another are halogen, C_1 - C_4 alkyl, C_2 - C_4 alkenyl, C_2 - C_4 alkynyl or C_1 - C_6 -alkoxy. Particular preference is given to compounds of the formula I in which R_2 is halogen, methyl, ethyl or ethinyl, and to compounds of the formula I in which R_1 and R_3 independently of one another are methyl, ethyl, isopropyl, vinyl, allyl, ethinyl, methoxy, ethoxy, bromine or chlorine. Very particular preference is given to compounds of the formula I in which G is the group - $C(X_1)$ - R_{30} or $C(X_2)$ - (X_3) - R_{31} in which X_1 , X_2 and X_3 are, in particular, oxygen, and R_{30} and R_{31} independently of one another are preferably C_1 - C_5 alkyl. Preference is furthermore given to compounds of the formula I in which R_{30} , R_{31} , R_{32} , R_{33} , R_{34} , R_{35} and R_{36} independently of one another are hydrogen, C_1 - C_5 alkyl or C_1 - C_5 haloalkyl.

Another preferred group of compounds of the formula I is is that where at least one ring atom of the groups Z_1 , Z_2 or Z_3 is bridged by an alkylene ring which, together with the carbon atoms of the groups Z_1 , Z_2 or Z_3 , contains 2 to 6 carbon atoms and may be interrupted by oxygen.

The invention also includes the salts which the compounds of the formula I can form with acids. Suitable acids for forming the acid addition salts are both organic and inorganic acids. Examples of such acids are hydrochloric acid, hydrobromic acid, nitric acid, phosphoric acids, sulfuric acid, acetic acid, propionic acid, butyric acid, valeric acid, oxalic acid, malonic acid, fumaric acid, organic sulfonic acids, lactic acid, tartaric acid, citric acid and salicylic acid. The salts of the compounds of the formula I with acidic hydrogen also include alkali metal salts, for example sodium salts and potassium salts; alkaline earth metal salts, for example calcium salts and magnesium salts; ammonium salts, i.e. unsubstituted ammonium salts and mono- or polysubstituted ammonium salts, and salts with other organic nitrogen bases. Correspondingly, suitable salt formers are alkali metal and alkaline earth metal hydroxides, in particular the hydroxides of lithium, sodium, potassium, magnesium or calcium, where those of sodium or potassium are particularly important.

Examples of amines which are suitable for forming ammonium salts are both ammonia and primary, secondary and tertiary C₁-C₁₈alkylamines, C₁-C₄hydroxyalkylamines and C₂-C4alkoxyalkylamines, for example methylamine, ethylamine, n-propylamine, isopropylamine, the four isomeric butylamines, n-amylamine, isoamylamine, hexylamine, heptylamine, octylamine, nonylamine, decylamine, pentadecylamine, hexadecylamine, heptadecylamine, octadecylamine, methylethylamine, methylisopropylamine, methylhexylamine, methylnonylamine, methylpentadecylamine, methyloctadecylamine, ethylbutylamine, ethylheptylamine, ethyloctylamine, hexylheptylamine, hexyloctylamine, dimethylamine, diethylamine, di-n-propylamine, diisopropylamine, di-n-butylamine, di-n-amylamine, diisoamylamine, dihexylamine, diheptylamine, dioctylamine, ethanolamine, npropanolamine, isopropanolamine, N,N-diethanolamine, N-ethylpropanolamine, Nbutylethanolamine, allylamine, n-butenyl-2-amine, n-pentenyl-2-amine, 2,3-dimethylbutenyl-2-amine, dibutenyl-2-amine, n-hexenyl-2-amine, propylenediamine, trimethylamine, triethylamine, tri-n-propylamine, triisopropylamine, tri-n-butylamine, triisobutylamine, tri-secbutylamine, tri-n-amylamine, methoxyethylamine and ethoxyethylamine; heterocyclic amines, for example pyridine, quinoline, isoquinoline, morpholine, N-methylmorpholine, thiomorpholine, piperidine, pyrrolidine, indoline, quinuclidine and azepine; primary arylamines, for example anilines, methoxyanilines, ethoxyanilines, o, m, p-toluidines, phenylenediamines, benzidines, naphthylamines and o, m, p-chloroanilines; but in particular triethylamine, isopropylamine and diisopropylamine.

In the processes described in this application, unless chiral starting materials are employed, the unsymmetrically substituted compounds of the formula I are generally obtained as racemates. The stereoisomers can then be separated by known methods, such as fractional crystallization after salt formation with optically pure bases, acids or metal complexes, or else by chromatographic processes such as high pressure liquid chromatography (HPLC) on acetylcellulose, owing to their physicochemical properties. In the present invention, the active compounds of the formula I are to be understood as meaning both the enriched and optically pure forms of the stereoisomers in question, and the racemates or diastereomers. Unless specific reference is made to the individual optical isomers, the given formula is to be understood as meaning those racemic mixtures which are formed in the preparation process mentioned. If an aliphatic C=C double bond is present, geometrical isomerism may additionally occur.

Also depending on the type of the substituents, the compounds of the formula I may be present as geometrical and/or optical isomers and isomer mixtures, and also as tautomers

and mixtures of tautomers. These compounds of the formula I likewise form part of the subject-matter of the present invention. The compounds of the formula I in which the group G is hydrogen may, for example, be present in the following tautomer equilibriums:

$$R_{5}$$
 R_{1}
 R_{2}
 R_{3}
 R_{4}
 R_{2}
 R_{3}
 R_{4}
 R_{5}
 R_{2}
 R_{4}
 R_{5}
 R_{4}
 R_{5}
 R_{5}
 R_{4}
 R_{5}
 R_{5}
 R_{4}
 R_{5}
 R_{5}
 R_{6}
 R_{7}
 R_{1}
 R_{2}
 R_{2}
 R_{3}
 R_{4}
 R_{5}
 R_{5}
 R_{4}
 R_{2}
 R_{4}
 R_{5}
 R_{5

If G is different from hydrogen and Z is the group Z_1 or Z_3 , or if G is different from hydrogen and Z_2 is unsymmetrically substituted, fused or spiro-linked, the compound of the formula I may be present as an isomer of the formula Id

Processes for preparing compounds which, with respect to the meaning of the substituents R_4 and R_5 , are different from the compounds of the formula I according to the present invention are, for example, described in WO 96/21652. The compounds of the formula I according to the present invention can be prepared by methods similar to the processes described in WO 96/21652. The compounds of the formula II

in which R_1 , R_2 , R_3 , R_4 and R_5 are as defined under formula I and which are employed as starting materials for such processes can be prepared, for example, by reacting a compound of the formula III

in which R is C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, preferably methyl, ethyl or trichloroethyl, and R_1 , R_2 and R_3 are as defined under formula I in an inert organic solvent, if appropriate in the presence of a base, with a compound of the formula IV or IVa

in which R_4 and R_5 are as defined under formula I. Other preparation processes for compounds of the formula II are described, for example, in WO 92/16510 .

The compounds of the formula III are either known, or they can be prepared similarly to known processes. Processes for preparing compounds of the formula III and their reaction with hydrazines are described, for example, in WO 97/02243 . Compounds of the formula III in which R is C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, preferably methyl, ethyl or trichloroethyl, and R_1 , R_2 and R_3 are as defined under formula I can be prepared by methods known to the person skilled in the art. For example, compounds of the formula III in which R is C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, preferably methyl, ethyl or trichloroethyl, and R_1 , R_2 and R_3 independently of

one another are C₁-C₄alkyl, C₂-C₄alkenyl, C₂-C₄alkynyl can be prepared by the process of cross-coupling according to Stille (J.K. Stille, Angew. Chem. <u>1986</u>, *98*, 504-519), Sonogashira (K. Sonogashira et al., Tetrahedron Lett. <u>1975</u>, 4467-4470), Suzuki (N. Miyaura, A. Suzuki, Chem. Rev. <u>1995</u>, *95*, 2457-2483) or Heck (R.F. Heck, Org. React. <u>1982</u>, *27*, 345-390), with or without subsequent hydrogenation. This procedure is illustrated by the following reaction scheme:

The compounds of the formulae IV and IVa are either known, or they can be prepared by known processes. Processes for preparing compounds of the formula IV are described, for example, in WO 95/00521. These compounds can be prepared, for example, by heating a compound of the formula V

$$R_{42}$$
 N
 R_{42}
 N
 R_{5}
 N

in which R_{42} is hydrogen, C_1 - C_4 alkyl, C_1 - C_6 alkoxy, C_1 - C_6 haloalkoxy, benzyloxy, preferably hydrogen, methyl, methoxy, ethoxy, trichloroethoxy, tert-butoxy or benzyloxy and R_4 and R_5 are as defined under formula I in the presence of a base or an acid in an inert solvent. Compounds of the formula V in which R_{42} is hydrogen, C_1 - C_4 alkyl, C_1 - C_6 alkoxy, C_1 - C_6 haloalkoxy, benzyloxy, preferably hydrogen, methyl, methoxy, ethoxy, trichloroethoxy, tert-butoxy or benzyloxy and R_4 and R_5 are as defined under formula I can be prepared, for example, by reacting a compound of the formula VI

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in which R₄₂ is hydrogen, C₁-C₄alkyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, benzyloxy, preferably hydrogen, methyl, methoxy, ethoxy, trichloroethoxy, tert-butoxy or benzyloxy in the presence of a base and an inert solvent with a compound of the formula VII

$$Y = Z_1, Z_2, \text{ or } Z_3 = Y \text{ (VII)},$$

in which Y is halogen, alkyl/aryl sulfonates -OSO₂R₄₃, preferably bromine, chlorine, iodine, mesylate ($R_{43}=CH_3$), triflate ($R_{43}=CF_3$) or tosylate ($R_{43}=p$ -tolyl) and Z_1 , Z_2 and Z_3 are as defined under formula I. In the formula VII, the free valencies of the groups Z_1, Z_2 and Z_3 are in each case attached to the group Y. Compounds of the formula VI and VII are known, or they can be prepared by methods known to the person skilled in the art.

Compounds of the formula IV in which R4 and R5 together are a group Z2 $-C-R_{14}(R_{15})-C-R_{16}(R_{17})-O-C-R_{18}(R_{19})-C-R_{20}(R_{21})-\\ (Z_2), in which \ R_{14},\ R_{15},\ R_{16},\ R_{17},\ R_{18},\ R_{19},\ R_{20}$ and R₂₁ are hydrogen can be prepared, for example, according to the following reaction scheme:

HO

$$CH_3SO_2O$$
 H_3C
 H_3

$$H_3C$$
 H_3C
 H_3C

Compounds of the formula I in which R_4 and R_5 are a group Z_1 or Z_3 can be prepared using the methods of the synthesis examples given above. Thus, the compounds of the formula III can, for example, be reacted with a hydrazine alkanol of the formula IV(b)

(here, R_{6} - R_{13} and R_{22} - R_{29} are hydrogen) to give the compounds of the formula IVc

followed by a cyclization, for example with formaldehyde, to give the end products of the formula le

The compound of the formula le in which R_1 and R_3 are ethyl and R_2 is methyl has a melting point of 186-191°C (decomp.). Similarly, it is also possible to prepare compounds of the formula I in which the substituents R_6 - R_{13} and R_{22} - R_{29} are different from hydrogen and, independently of one another, have one of the meanings mentioned for them above.

The end products of the formula I can be isolated in a customary manner by concentration and/or evaporation of the solvent and be purified by recrystallization or trituration of the solid residue in solvents in which they are not readily soluble, such as ethers, alkanes, aromatic hydrocarbons or chlorinated hydrocarbons, or by chromatography. Salts of compounds of the formula I can be prepared in a manner known per se. Such preparation methods are described, for example, in WO 96/21652.

The compounds of the formula I or compositions comprising them can be used according to the invention by all the application methods customary in agriculture, for example pre-emergence application, postemergence application and seed dressing, and various methods and techniques, for example controlled release of active compounds. To this end, the active compound is absorbed in solution onto mineral granule carriers or polymerized granules (urea/formaldehyde) and dried. If appropriate, a coating which allows the active compound to be released in metered form over a certain period of time can additionally be applied (coated granules).

The compounds of the formula I can be employed as herbicides in unchanged form, i.e. as they are obtained in the synthesis, but they are preferably processed in a customary manner with the auxiliaries conventionally used in the art of formulation, for example to give emulsifiable concentrates, directly sprayable or dilutable solutions, dilute emulsions, wettable powders, soluble powders, dusts, granules or microcapsules. Such formulations are described, for example, in WO 97/34485 on pages 9 to 13. The methods of application, such as spraying, atomizing, dusting, wetting, scattering or watering, in the same way as the nature of the compositions, are chosen according to the required aims and the given circumstances.

The formulations, i.e. the compositions, formulations or preparations comprising the active compound of the formula I or at least one active compound of the formula I and as a rule one or more solid or liquid formulation auxiliaries, are prepared in a known manner, for example by intimate mixing and/or grinding of the active compounds with the formulation auxiliaries, for example solvents or solid carriers. Surface-active compounds (surfactants) can furthermore additionally be used during the preparation of the formulations. Examples of solvents and solid carriers are given, for example, in WO 97/34485 on page 6. Depending on the nature of the active compound of the formula I to be formulated, suitable surface-active compounds are nonionic, cationic and/or anionic surfactants and surfactant mixtures having good emulsifying, dispersing and wetting properties.

Examples of suitable anionic, nonionic and cationic surfactants are listed, for example, in WO 97/34485 on pages 7 and 8.

The surfactants conventionally used in the art of formulation and which can also be used to prepare the herbicidal compositions according to the invention are described, inter alia, in "Mc Cutcheon's Detergents and Emulsifiers Annual", MC Publishing Corp., Ridgewood New Jersey, 1981, Stache, H., "Tensid-Taschenbuch" [Surfactant handbook], Carl Hanser Verlag, Munich/Vienna, 1981 and M. and J. Ash, "Encyclopedia of Surfactants", Vol I-III, Chemical Publishing Co., New York, 1980-81.

The efficacy of herbicidal and plant-growth-inhibiting compositions according to the invention containing a herbicidally effective amount of a compound of the formula I can be enhanced by addition of spray tank adjuvants.

These adjuvants may be, for example: nonionic surfactants, mixtures of nonionic surfactants, mixtures of anionic surfactants with nonionic surfactants, cationic surfactants, organosilicon surfactants, mineral oil derivatives with and without surfactants, vegetable oil derivatives with and without addition of surfactants, alkylated derivatives of oils of vegetable or mineral origin with and without surfactants, fish oils and other oils of animal nature and their alkyl derivatives with and without surfactants, natural higher fatty acids, preferably having 8 to 28 carbon atoms, and their alkyl ester derivatives, organic acids which contain an aromatic ring system and one or more carboxylic esters, and their alkyl derivatives, furthermore suspensions of polymers of vinyl acetate or copolymers of vinyl acetate/acrylic esters. Mixtures of individual adjuvants with one another and in combination with organic solvents may further increase the effect.

Suitable nonionic surfactants are, for example, polyglycol ether derivatives of aliphatic or cycloaliphatic alcohols, saturated or unsaturated fatty acids and alkylphenols, preferably those which may contain 3 to 30 glycol ether groups and 8 to 20 carbon atoms in the (aliphatic) hydrocarbon radical and 6 to 18 carbon atoms in the alkyl radical of the alkylphenols.

Other suitable nonionic surfactants are the water-soluble polyethylene oxide adducts on polypropylene glycol, ethylenediaminopolypropylene glycol and alkylpolypropylene glycol preferably having 1 to 10 carbon atoms in the alkyl chain which preferably contain 20 to 250 ethylene glycol ether groups and 10 to 100 propylene glycol ether groups. The abovementioned compounds generally contain 1 to 5 ethylene glycol units per propylene glycol unit.

Other examples of nonionic surfactants which may be mentioned are nonylphenolpolyethoxyethanols, castor oil polyglycol ethers, polypropylene/polyethylene oxide adducts, tributylphenoxypolyethoxyethanol, polyethylene glycol and octylphenoxypolyethoxyethanol.

Also suitable are fatty esters of polyoxyethylene sorbitan, for example polyoxyethylene sorbitan trioleate.

Preferred anionic surfactants are, in particular, alkyl sulfates, alkyl sulfonates, alkylaryl sulfonates, alkylated phosphoric acids and their ethoxylated derivatives. The alkyl radicals usually contain 8 to 24 carbon atoms.

Preferred nonionic surfactants are known under the following trade names:

Polyoxyethylene cocoalkylamine (for example AMIET® 105 (Kao Co.)), polyoxyethylene oleylamine (for example AMIET® 415 (Kao Co.)), nonylphenolpolyethoxyethanols, polyoxyethylene stearylamine (for example AMIET® 320 (Kao Co.)), N-polyethoxyethylamines (for example GENAMIN® (Hoechst AG)), N,N,N',N'-tetra(polyethoxypolypropoxyethyl)ethylene diamines (for example TERRONIL® and TETRONIC® (BASF Wyandotte Corp.)), BRIJ® (Atlas Chemicals), ETHYLAN® CD and ETHYLAN® D (Diamond Shamrock), GENAPOL® C, GENAPOL® O, GENAPOL® S and GENAPOL® X080 (Hoechst AG), EMULGEN® 104P, EMULGEN® 109P and EMULGEN® 408 (Kao Co.); DISTY® 125 (Geronazzo), SOPROPHOR® CY 18 (Rhône Poulenc S.A.); NONISOL® (Ciba-Geigy), MRYJ® (ICI); TWEEN® (ICI); EMULSOGEN® (Hoechst AG); AMIDOX® (Stephan Chemical Co.), ETHOMID® (Armak Co.); PLURONIC® (BASF Wyandotte Corp.), SOPROPHOR® 461P (Rhône Poulenc S.A.), SOPROPHOR® 496/P (Rhône Poulenc S.A.), ANTAROX FM-63 (Rhône Poulenc S.A.), SLYGARD 309 (Dow Corning), SILWET 408, SILWET L-7607N (Osi-Specialities).

The cationic surfactants are primarily quaternary ammonium salts which contain, as N-substituents, at least one alkyl radical having 8 to 22 C atoms and, as further substituents, lower nonhalogenated or halogenated alkyl, benzyl or lower hydroxyalkyl radicals. The salts are preferably present as halides, methyl sulfates or ethyl sulfates, for example stearyltrimethylammonium chloride or benzyldi(2-chloroethyl)ethylammonium bromide.

The oils used are either of mineral or natural origin. The natural oils may additionally be of animal or vegetable origin. In the case of animal oils, preference is given, in particular, to derivatives of beef tallow, but fish oils (for example sardine oil) and derivatives thereof are also used. Vegetable oils are mainly seed oils of various origin. Examples of particularly preferred vegetable oils which may be mentioned are coconut, rapeseed or sunflower oils and derivatives thereof.

Surfactants, oils, in particlar vegetable oils, derivatives thereof such as alkylated fatty acids and mixtures thereof, for example with preferably anionic surfactants such as alkylated phosphoric acids, alkyl sulfates and alkylaryl sulfonates and higher fatty acids which are customary in formulation and adjuvant technique and which can also be employed in the compositions according to the invention and spray tank solutions thereof are described,

inter alia, in "Mc Cutcheon's Detergents and Emulsifiers Annual", MC Publishing Corp., Ridgewood New Jersey, 1998, Stache, H., "Tensid-Taschenbuch" [Surfactant handbook], Carl Hanser Verlag, Munich/Vienna, 1990, M. and J. Ash, "Encyclopedia of Surfactants", Vol. I-IV, Chemical Publishing Co., New York, 1981-89, G. Kapusta, "A Compendium of Herbicide Adjuvants", Southern Illinois Univ., 1998, L. Thomson Harvey, "A Guide to Agricultural Spray Adjuvants Used in the United States", Thomson Pubns., 1992.

The herbicidal formulations as a rule comprise 0.1 to 99% by weight, in particular 0.1 to 95% by weight, of herbicide, 1 to 99.9% by weight, in particular 5 to 99.8% by weight, of a solid or liquid formulation auxiliary and 0 to 25% by weight, in particular 0.1 to 25% by weight, of a surfactant. While concentrated compositions are rather preferred as commercial goods, the end user as a rule uses dilute compositions. The compositions can also comprise further additives, such as stabilizers, for example epoxidized or non-epoxidized vegetable oils (epoxidized coconut oil, rapeseed oil or soya oil), defoamers, for example silicone oil, preservatives, viscosity regulators, binders, tackifiers and fertilizers or other active compounds.

The herbicidally active compounds of the formula I are as a rule applied to the plants or their habitat, at application rates of 0.001 to 4 kg/ha, in particular 0.005 to 2 kg/ha. The dosage required for the desired effect can be determined by tests. It depends on the nature of the effect, the development stage of the crop plant and the weed and on the application (location, time, process) and can, as a function of these parameters, vary within wide ranges.

The compounds of the formula I have herbicidal and growth-inhibiting properties, owing to which they can be used in crops of useful plants, in particular in cereals, cotton, soya, sugar beet, sugar cane, plantings, rapeseed, maize and rice, very particularly in maize and cereals, and for the non-selective control of weeds. Crops include those which have been rendered tolerant towards herbicides or herbicide classes by conventional breeding methods or genetical engineering methods. The weeds to be controlled can be both monocotyledonous and dicotyledonous weeds, for example Stellaria, Agrostis, Digitaria, Avena, Brachiaria, Phalaris, Setaria, Sinapis, Lolium, Solanum, Echinochloa, Scirpus, Monochoria, Sagittaria, Panicum, Bromus, Alopecurus, Sorghum halepense, Sorghum bicolor, Rottboellia, Cyperus, Abutilon, Sida, Xanthium, Amaranthus, Chenopodium, Ipomoea, Chrysanthemum, Galium, Viola, Matricharia, Papaver and Veronica. The herbicidal composition according to the invention is particularly suitable for controlling

Alopecurus, Avena, Agrostis, Setaria, Phalaris, Lolium, Panicum, Echinochloa, Brachiaria and Digitaria.

Surprisingly, it has been found that specific safeners known from US-A-5 041 157, US-A-5-541 148, US-A-5 006 656, EP-A-0 094 349, EP-A-0 551 650, EP-A-0 268 554, EP-A-0 375 061, EP-A-0 174 562, EP-A-492 366, WO 91/7874, WO 94/987, DE-A-19 612 943, WO 96/29870, WO 98/13361, WO 98/39297, WO 98/27049, EP 716 073, EP 613 618, US-A-5 597 776 and EP-A-430 004 are suitable for mixing with the herbicidal composition according to the invention. Consequently, the present invention also relates to a selective herbicidal composition for controlling grasses and weeds in crops of useful plants, in particular in crops of maize and cereals, said composition comprising a herbicide of the formula I and a safener (antidote) and which protects the useful plants, but not the weeds, against the phytotoxic effect of the herbicide, and to the use of this composition for controlling weeds in crops of useful plants.

According to the invention, a selective-herbicidal composition is therefore proposed which, in addition to customary inert formulation auxiliaries such as carriers, solvents and wetting agents, comprises, as active compound, a mixture of

a) a herbicidally effective amount of a compound of the formula I

in which R_1 , R_2 , R_3 , R_4 , R_5 and G are as defined above, and

b) a herbicide-antagonistically effective amount of either a compound of the formula X

$$X_6$$
 $O - CH_2 - O - R_{37}$
 $O - CH_2 - O - R_{37}$

in which R_{37} is hydrogen, C_1 - C_6 alkyl or C_1 - C_6 alkoxy- or C_3 - C_6 alkenyloxy-substituted C_1 - C_6 alkyl; and X_6 is hydrogen or chlorine; or a compound of the formula X_1

in which

E is nitrogen or methine;

R₃₈ is -CCl₃, phenyl or halogen-substituted phenyl;

 R_{39} and R_{40} independently of one another are hydrogen or halogen; and R_{41} is C_1 - C_4 alkyl; or a compound of the formula XII

$$R_{47}O_2C$$
 N
 R_{46}
 R_{45}
 R_{44}
 R_{44}
 R_{44}
 R_{44}
 R_{45}

in which R_{44} and R_{45} independently of one another are hydrogen or halogen and R_{46} , R_{47} and R_{48} independently of one another are C_1 - C_4 alkyl, or a compound of the formula XIII

$$R_{51}$$
 N -CO-N
 R_{52}
 R_{53}
 R_{53}
 R_{50}
 R_{50}
 R_{50}
 R_{50}
 R_{50}
 R_{51}
 R_{50}
 R_{50}
 R_{50}
 R_{50}
 R_{50}
 R_{50}
 R_{50}
 R_{50}

in which A2 is a group

R₅₁ and R₅₂ independently of one another are hydrogen, C₁-C₈alkyl, C₃-C₈cycloalkyl,

$$C_3$$
- C_6 alkenyl, C_3 - C_6 alkynyl, R_y , or C_1 - C_4 alkoxy- or R_y

substituted C_1 - C_4 alkyl; or R_{51} and R_{52} together form a C_4 - C_6 alkylene bridge which may be interrupted by oxygen, sulfur, SO, SO₂, NH or -N(C_1 - C_4 alkyl)-,

R₅₃ is hydrogen or C₁-C₄alkyl;

 R_{49} is hydrogen, halogen, cyano, trifluoromethyl, nitro, C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfinyl, -COOR_j, -CONR_kR_m, -COR_n, -SO₂NR_kR_m or -OSO₂- C_1 - C_4 alkyl;

 R_g is hydrogen, halogen, cyano, nitro, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, -COOR_j, -CONR_kR_m, -COR_n, -SO₂NR_kR_m, -OSO₂- C_1 - C_4 alkyl, C_1 - C_6 alkoxy, or C_1 - C_6 alkoxy which is substituted by C_1 - C_4 alkoxy or halogen, C_3 - C_6 alkenyloxy, or C_3 - C_6 alkenyloxy which is substituted by halogen, or C_3 - C_6 alkynyloxy, or C_4 - C_4 alkylene bridge which may be substituted by halogen or C_1 - C_4 alkyl, or they form a C_3 - C_4 alkenylene bridge which may be substituted by halogen or C_1 - C_4 alkyl, or they form a C_4 alkadienylene bridge which may be substituted by halogen or C_1 - C_4 alkyl;

 R_{50} and R_h independently of one another are hydrogen, halogen, C_1 - C_4 alkyl, trifluoromethyl, C_1 - C_6 alkoxy, C_1 - C_6 alkylthio or -COOR;

 R_c is hydrogen, halogen, nitro, C_1 - C_4 alkyl or methoxy; R_d is hydrogen, halogen, nitro, C_1 - C_4 alkyl, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, -COOR_j or CONR_kR_m:

 R_e is hydrogen, halogen, C_1 - C_4 alkyl, -COOR $_j$, trifluoromethyl or methoxy, or R_d and R_e together form a C_3 - C_4 alkylene bridge;

Rp is hydrogen, halogen, C₁-C₄alkyl, -COOR_j, trifluoromethyl or methoxy; Rq is hydrogen, halogen, nitro, C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, -COOR_j or CONR_kR_m, or Rp and Rq together form a C₃-C₄alkylene bridge;

Rr is hydrogen, halogen, C₁-C₄alkyl, -COOR_j, trifluoromethyl or methoxy; Rs is hydrogen, halogen, nitro, C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, -COOR_j or CONR_kR_m, or Rr and Rs together form a C₃-C₄alkylene bridge;

Rt is hydrogen, halogen, C₁-C₄alkyl, -COOR_j, trifluoromethyl or methoxy; Ru is hydrogen, halogen, nitro, C₁-C₄alkyl, C₁-C₄alkyl, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfinyl, -COOR_j or CONR_kR_m, or Rv and Ru together form a C₃-C₄alkylene bridge;

R_f and Rv are hydrogen, halogen or C₁-C₄alkyl;

 R_x and R_y independently of one another are hydrogen, halogen, C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylthio, -COOR $_{54}$, trifluoromethyl, nitro or cyano;

 R_j , R_k and R_m independently of one another are hydrogen or C_1 - C_4 alkyl; or R_k and R_m together form a C_4 - C_6 alkylene bridge which may be interrupted by oxygen, NH or -N(C_1 - C_4 alkyl)-;

 R_n is C_1 - C_4 alkyl, phenyl, or halogen-, C_1 - C_4 alkyl-, methoxy-, nitro- or trifluoromethyl-substituted phenyl;

 $R_{54} \text{ is hydrogen, } C_1\text{-}C_1\text{-}alkyl, C_1\text{-}C_4\text{alkoxy-}C_1\text{-}C_4\text{alkyl, } C_1\text{-}C_4\text{alkylthio-}C_1\text{-}C_4\text{alkyl, } \text{di-}C_1\text{-}C_4\text{alkylamino-}C_1\text{-}C_4\text{alkyl, } \text{halo-}C_1\text{-}C_8\text{alkyl, } C_2\text{-}C_8\text{alkenyl, } \text{halo-}C_2\text{-}C_8\text{alkenyl, } C_3\text{-}C_7\text{cycloalkyl, } \text{halo-}C_3\text{-}C_7\text{cycloalkyl, } C_1\text{-}C_8\text{alkylcarbonyl, } \text{allylcarbonyl, } C_3\text{-}C_7\text{cycloalkyl, } C_1\text{-}C_8\text{alkylcarbonyl, } C_3\text{-}C_8\text{alkylcarbonyl, } C_3\text{-}C_8\text{$

 C_7 cycloalkylcarbonyl, benzoyl which is unsubstituted or substituted up to three times on the phenyl ring by identical or different substituents selected from the group consisting of halogen, C_1 - C_4 alkyl, halo- C_1 - C_4 alkoxy or C_1 - C_4 alkoxy; or furoyl, thienyl; or C_1 - C_4 alkyl which is substituted by phenyl, halophenyl, C_1 - C_4 alkylphenyl, C_1 - C_4 alkoxyphenyl, halo- C_1 - C_4 alkylphenyl, halo- C_1 - C_4 alkoxyphenyl, C_1 - C_6 alkoxycarbonyl, C_1 - C_4 alkoxy- C_1 - C_6 alkoxycarbonyl, C_3 - C_8 alkenyloxycarbonyl, C_3 - C_8 alkenyloxycarbonyl, C_3 - C_8 alkenyloxycarbonyl, C_3 - C_8 alkynyloxycarbonyl, C_1 -

 C_8 alkylthiocarbonyl, C_3 - C_8 alkenylthiocarbonyl, C_3 - C_8 alkynylthiocarbonyl, carbamoyl, mono- C_1 - C_4 alkylaminocarbonyl, di- C_1 - C_4 alkylaminocarbonyl; or phenylaminocarbonyl which is unsubstituted or substituted up to three times on the phenyl by identical or different substituents selected from the group consisting of halogen, C_1 - C_4 alkyl, halo- C_1 - C_4 alkyl, halo- C_1 - C_4 alkoxy and C_1 - C_4 alkoxy, or is monosubstituted by cyano or nitro, or dioxolan-2-yl which is unsubstituted or substituted by one or two C_1 - C_4 alkyl radicals, or dioxan-2-yl which is unsubstituted or substituted by one or two C_1 - C_4 alkyl radicals, or C_1 - C_4 alkyl which is substituted by cyano, nitro, carboxyl or C_1 - C_8 alkylthio- C_1 - C_8 alkoxycarbonyl; or a compound of the formula XIV

$$R_{56}$$
 N $CHCl_2$ (XIV),

in which R_{56} and R_{57} independently of one another are $C_1\text{-}C_6 alkyl$ or $C_2\text{-}C_6 alkenyl;$ or R_{56} and

$$R_{57}$$
 together are ; R_{58} and R_{59} independently of one another are hydrogen

or
$$C_1$$
- C_6 alkyl; or R_{56} and R_{57} together are R_{60}

 R_{62} is hydrogen, $C_1\text{-}C_4alkyl\ or$

 R_{60} and R_{61} independently of one another are C_1 - C_4 alkyl, or R_{60} and R_{61} together are -(CH_2)₅-;

or
$$R_{56}$$
 and R_{57} together are R_{69} R_{69} R_{69} R_{70} R_{76} R_{76} R_{77} R_{78}

 R_{63} , R_{64} , R_{65} , R_{66} , R_{67} , R_{68} , R_{69} , R_{70} , R_{71} , R_{72} , R_{73} , R_{74} , R_{75} , R_{76} , R_{77} and R_{78} independently of one another are hydrogen or C_1 - C_4 alkyl; or a compound of the formula XV

$$R_{80}$$
 $N-O$
 O
 O
 (XV)

in which R_{80} is hydrogen or chlorine and R_{79} is cyano or trifluoromethyl, or a compound of the formula XVI

in which R₈₁ is hydrogen or methyl, or of the formula XVII

$$R_{83}$$

$$R_{84}$$

$$V_{1}$$

$$V_{1}$$

$$V_{1}$$

$$(XVII),$$

in which

 R_{82} is hydrogen, C_1 - C_4 alkyl, C_1 - C_4 alkyl which is substituted by C_1 - C_4 alkyl- X_2 - or C_1 - C_4 haloalkyl- X_2 -, C_1 - C_4 haloalkyl, nitro, cyano, -COOR₈₅, -NR₈₆R₈₇, -SO₂NR₈₈R₈₉ or -CONR₉₀R₉₁;

 R_{83} is hydrogen, halogen, C_1 - C_4 alkyl, trifluoromethyl, C_1 - C_4 alkoxy or C_1 - C_4 haloalkoxy; R_{84} is hydrogen, halogen or C_1 - C_4 alkyl;

U, V, W_1 and Z_4 independently of one another are oxygen, sulfur, $C(R_{92})R_{93}$, carbonyl, NR_{94} ,

a group
$$C = \bigcap_{R_{95}} \bigcap_{R_{96}} \bigcap_{R_{96}} C = C \cap_{R_{95}} \bigcap_{R_{96}} \bigcap_$$

C₂-C₄alkynyl; with the proviso that

a) at least one of the ring members U, V, W₁ or Z₄ is carbonyl, and a ring member which is

adjacent to this or these ring members is the group

b) two adjacent ring members U and V, V and W_1 and W_2 and W_3 and W_4 may not simultaneously be oxygen;

R₉₅ and R₉₆ independently of one another are hydrogen or C₁-C₈alkyl; or

R₉₅ and R₉₆ together form a C₂-C₆alkylene group;

 A_1 is R_{99} - Y_1 - or - $NR_{97}R_{98}$;

X₂ is oxygen or -S(O)_s;

Y₁ is oxygen or sulfur;

 R_{99} is hydrogen, C_1 - C_8 alkyl, C_1 - C_8 haloalkyl, C_1 - C_4 alkoxy- C_1 - C_8 alkyl, C_3 - C_6 alkenyloxy- C_1 - C_8 alkyl or phenyl- C_1 - C_8 alkyl, where the phenyl ring may be substituted by halogen, C_1 - C_4 alkyl, trifluoromethyl, methoxy or methyl- $S(O)_5$ -, C_3 - C_6 alkenyl, C_3 - C_6 haloalkenyl, phenyl- C_3 - C_6 alkenyl, C_3 - C_6 alkynyl, phenyl- C_3 - C_6 alkynyl, oxetanyl, furyl or tetrahydrofuryl;

R₈₅ is hydrogen or C₁-C₄alkyl;

R₈₆ is hydrogen, C₁-C₄alkyl or C₁-C₄alkylcarbonyl;

R₈₇ is hydrogen or C₁-C₄alkyl; or

R₈₆ and R₈₇ together form a C₄- or C₅alkylene group;

 R_{88} , R_{89} , R_{90} and R_{91} independently of one another are hydrogen or C_1 - C_4 alkyl; or R_{88} together with R_{89} or R_{90} together with R_{91} independently of one another are C_4 - or C_5 -alkylene, where a carbon atom may be replaced by oxygen or sulfur, or one or two carbon atoms may be replaced by -NR₁₀₀-;

 R_{92} , R_{100} and R_{93} independently of one another are hydrogen or C_1 - C_8 alkyl; or R_{92} and R_{93} together are C_2 - C_6 alkylene;

R₉₄ is hydrogen or C₁-C₈alkyl;

 R_{97} is hydrogen, C_1 - C_8 alkyl, phenyl, phenyl- C_1 - C_8 alkyl, where the phenyl rings may be substituted by fluorine, chlorine, bromine, nitro, cyano, -OCH₃, C_1 - C_4 alkyl or CH₃SO₂-, C_1 - C_4 alkoxy- C_1 - C_8 alkyl, C_3 - C_6 alkenyl or C_3 - C_6 alkynyl;

R₉₈ is hydrogen, C₁-C₈alkyl, C₃-C₆alkenyl or C₃-C₆alkynyl; or

 R_{97} and R_{98} together are C_4 - or C_5 -alkylene, where a carbon atom may be replaced by oxygen or sulfur, or one or two carbon atoms may be replaced by -NR₁₀₁-; R_{101} is hydrogen or C_1 - C_4 alkyl;

r is 0 or 1; and

s is 0, 1 or 2, or a compound of the formula XVIII

$$\begin{array}{c|c}
 & N \\
 & N \\$$

in which R_{103} is hydrogen, C_1 - C_6 alkyl, C_3 - C_6 cycloalkyl, C_3 - C_6 alkenyl or C_3 - C_6 alkynyl; and R_{104} , R_{105} and R_{106} independently of one another are hydrogen, C_1 - C_6 alkyl, C_3 - C_6 cycloalkyl or C_1 - C_6 alkoxy, with the proviso that one of the substituents R_{104} , R_{105} and R_{106} is different from hydrogen;

a compound of the formula XIX

in which Z_5 is N or CH, n, in the case where Z_5 is N, is 0, 1, 2 or 3 and, in the case where Z_5 is CH, is 0, 1, 2, 3 or 4, R_{107} is halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy, nitro, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl, C_1 - C_4 alkoxycarbonyl or unsubstituted or substituted phenyl or phenoxy, R_{108} is hydrogen or C_1 - C_4 alkyl, R_{109} is hydrogen, C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, C_2 - C_6 alkenyl, C_2 - C_6 alkynyl, C_1 - C_4 alkyl, C_1 - C_4 alkyl, C_1 - C_4 alkylthio- C_1 - C_4 alkyl, C_1 - C_4 alkylsulfonyl- C_1 - C_4 alkyl, C_1 -C

in which Z₆ is O or N-R₁₁₀ and R₁₁₀ is a group of the formula

in which R_{111} and R_{112} independently of one another are cyano, hydrogen, C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, C_2 - C_6 alkenyl, unsubstituted or substituted phenyl or heteroaryl;

a compound of the formula XXI
$$Z_7$$
 R_{114} (XXI) ,

in which Z_7 is O, S, S=O, SO₂ or CH₂, R₁₁₃ and R₁₁₄ independently of one another are hydrogen, halogen or C₁-C₄alkyl, W₂ and W₃ independently of one another are CH₂COOR₁₁₅, COOR₁₁₅ or together are a group of the formula -(CH₂)C(O)-O-C(O)-(CH₂)-, and R₁₁₅ is hydrogen, C₁-C₄alkyl, C₂-C₄alkenyl, C₂-C₆alkynyl, C₃-C₆cycloalkyl, C₁-C₄haloalkyl, a metal cation or an ammonium cation; a compound of the formula XXII

$$R_{120} \longrightarrow Z_8 \longrightarrow W_4 \longrightarrow (XXII),$$

in which R_{119} and R_{120} independently of one another are hydrogen, halogen or C_1 - C_4 haloalkyl, R_{121} is hydrogen, C_1 - C_4 alkyl, C_3 - C_4 alkenyl, C_3 - C_4 alkynyl, C_1 - C_4 haloalkyl, C_3 - C_6 cycloalkyl, a metal cation or an ammonium cation, Z_8 is N, CH, C-F or C-Cl and W_4 is a group of the formula

in which R_{122} and R_{123} independently of one another are hydrogen or C_1 - C_4 alkyl and R_{124} and R_{125} independently of one another are hydrogen or C_1 - C_4 alkyl; a compound of the formula XXIII

in which R_{126} is hydrogen, cyano, halogen, C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkoxycarbonyl, C_1 - C_4 alkylthiocarbonyl, -NH- R_{128} , -C(O)NH- R_{128} , unsubstituted or substituted aryl or heteroaryl,

 R_{127} is hydrogen, cyano, nitro, halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 thioalkyl, C_1 - C_4 haloalkyl, -NH- R_{128} , -C(O)NH- R_{128} , unsubstituted or substituted aryl, heteroaryl, and R_{128} is C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_3 - C_4 alkenyl, C_3 - C_4 alkynyl, C_3 - C_4 cycloalkyl, unsubstituted or substituted aryl or heteroaryl, formyl, C_1 - C_4 -alkylcarbonyl, C_1 - C_4 -alkylsulfonyl;

a compound of the formula XXIV

$$R_{132}$$
 R_{131}
 R_{130}
 R_{132}
 R_{133}
 R_{129}
 R_{129}
 R_{129}

in which R_{129} and R_{130} independently of one another are hydrogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, mono- C_1 - C_8 - or di- C_1 - C_8 alkylamino, C_3 - C_6 cycloalkyl, C_1 - C_4 thioalkyl, phenyl or heteroaryl, R_{131} has the meaning of R_{129} and is additionally OH, NH_2 , halogen, di- C_1 - C_4 aminoalkyl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl or C_1 - C_4 alkoxycarbonyl, R_{132} has the meaning of R_{129} and is additionally cyano, nitro, carboxyl, C_1 - C_4 alkoxycarbonyl, di- C_1 - C_4 aminoalkyl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl, SO_2 -OH, iso- C_1 - C_2 -alkylsulfonyl or C_1 - C_3 -alkoxyculfonyl, C_4 - C_4 -alkylsulfonyl, C_1 - C_4 - C_4 -alkylsulfonyl, C_1 - C_4

iso- C_1 - C_4 aminoalkylsulfonyl or C_1 - C_4 alkoxysulfonyl, R_{133} has the meaning of R_{129} and is additionally OH, NH₂, halogen, di- C_1 - C_4 aminoalkyl, pyrrolidin-1-yl, piperidin-1-yl,

morpholin-1-yl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl, C_1 - C_4 alkoxycarbonyl, phenoxy, naphtoxy, phenylamino, benzoyloxy or phenylsulfonyloxy; or a compound of the formula XXV

in which R_{134} is hydrogen, C_4 alkyl, C_1 - C_4 haloalkyl, C_2 - C_4 alkenyl, C_2 - C_4 alkynyl or C_1 - C_4 alkoxy- C_1 - C_4 alkyl, R_{135} is hydrogen, halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or C_1 - C_4 alkoxy and R_{136} is hydrogen, halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or C_1 - C_4 alkoxy, with the proviso that R_{135} and R_{136} are not simultaneously hydrogen.

The selective-herbicidal composition according to the invention preferably comprises, as herbicide-antagonistically effective amount, either a compound of the formula X

$$\begin{array}{c}
X_6 \\
O - CH_2 \longrightarrow O - R_{37}
\end{array}$$
(X),

in which R_{37} is hydrogen, C_1 - C_8 alkyl or C_1 - C_6 alkoxy- or C_3 - C_6 alkenyloxy-substituted C_1 - C_8 alkyl; and X_6 is hydrogen or chlorine; or a compound of the formula XI

in which

E is nitrogen or methine; R₃₈ is -CCl₃, phenyl or halogen-substituted phenyl;

 R_{39} and R_{40} independently of one another are hydrogen or halogen; and R_{41} is C_1 - C_4 alkyl; or a compound of the formula XII

in which R_{44} and R_{45} independently of one another are hydrogen or halogen and R_{46} , R_{47} and R_{48} independently of one another are C_1 - C_4 alkyl.

The abovementioned preferences for the compounds of the formula I also apply to mixtures of the compounds of the formula I with safeners of the formulae X to XVIII. Preferred compositions according to the invention comprise a safener selected from the group consisting of the formula Xa

$$CI$$

$$O-CH_2-C(O)-O-CH(CH_3)C_5H_{11}-n$$
(Xa),

the formula Xb

and the formula XIa

Other preferred compounds of the formulae X, XI and XII are also listed in Tables 9, 10 and 11.

Table 9: Compounds of the formula X:

$$\begin{array}{c}
X_6 \\
N \\
O - CH_2 \\
O - R_{37}
\end{array}$$
(X),

Comp. No. X₆ R₃₇

9.01	Cl	-CH(CH₃)-C₅H₁₁-n
9.02	Cl	-CH(CH ₃)-CH ₂ OCH ₂ CH=CH ₂
9.03	CI	Н
9.04	Cl	C₄H ₉ -n

Preferred compounds of the formula XI are listed in Table 10 below.

Table 10: Compounds of the formula XI:

Comp. No.	R ₄₁	R ₃₈	R ₃₉	R ₄₀		E
						,
10.01	CH₃	phenyl		2-CI	Н	СН
10.02	CH₃	phenyl		2-CI	4-CI	СН
10.03	CH₃	phenyl		2-F	Н	СН
10.04	CH₃	2-chlore	phenyl	2-F	Н	СН
10.05	C₂H	5 CCI ₃		2-CI	4-CI	N
10.06	CH₃	phenyl		2-CI	4-CF ₃	Ν
10.07	CH ₃	phenyl		2-CI	4-CF ₃	N

Preferred compounds of the formula XII are listed in Table 11 below.

Table 11: Compounds of the formula XII:

$$R_{47}O_2C$$
 R_{46}
 R_{45}
 R_{44}
 R_{44}
 R_{45}

Comp. No.	R ₄₆	R ₄₇	R ₄₈	R ₄₄	R ₄₅
11.01	CH ₃	CH ₃	CH ₃	2-Cl	4-CI
11.02	CH₃	C ₂ H ₅	CH ₃	2-Cl 2-Cl	4-Cl
11.03	СНз	C ₂ H ₅	C₂H₅	2-Cl	4-CI

Preferred compounds of the formula XIII are listed in Table 12 below as compounds of the formula XIIIa:

Table 12: Compounds of the formula XIIIa:

$$A_{2} \longrightarrow NHSO_{2} \longrightarrow N$$

Preferred compounds of the formula XIV are listed in Table 13 below:

Table 13: Compounds of the formula XIV:

$$R_{56}$$
 N $CHCl_2$ (XIV)

Comp. No.	R ₅₆	R ₅₇	R ₅₆ +R ₅₇
13.001	CH ₂ =CHCH ₂	CH ₂ =CHCH ₂	-
13.002			CH ₃ C CH ₃
13.003			сн, сн,
13.004	-	-	

Preferred compounds of the formula XV are listed in Table 14 below:

Table 14: Compounds of the formula XV:

13.008

$$R_{80} \longrightarrow R_{79} \qquad O \qquad (XV)$$

Comp. No.	R ₈₀	R ₇₉
14.01	Н	CN
14.02	Cl	CF ₃

Preferred compounds of the formula XVI are listed in Table 15 below:

Table 15: Compounds of the formula XVI:

$$R_{g_1} \longrightarrow N \longrightarrow CI$$

$$N \longrightarrow CI$$

$$N \longrightarrow CI$$

$$CI$$

	Cl
Comp. No.	R ₈₁
15.01	Н
15.02	CH₃

Preferred compounds of the formula XVII are listed in Table 16 below as compounds of the formula XVIIa:

Table 16: Compounds of the formula XVIIa

$$R_{82}$$
 $(V)_r$ $(XVIIa)$

Comp. No.	R ₈₂	Z_4	V	r
16.001	Н	C = C C C C C C C C C C	0	1
16.002	Н	C=CH COOCH ₃	0	1
16.003	Н	C=CH C C CH	0	1
16.004	Н	C=CH	0	1
16.005	Н	C=CH COOCH ₃	CH ₂	1
16.006	Н	C=CH CH ³ CH ³	CH ₂	1
16.007	Н	C=CH COOCH ₃	S	1
16.008	н	C=CH O_CH ₂	S	1
16.009	Н	C=CH_CFCH	NCH₃	1
16.010	Н	C=CH COOCH3	NCH ₃	1
16.011	Н	C=CH CH ³	NCH₃	1
16.012	Н	C=CH CH ³	0	1
16.013	Н	C=CH CH ³ CH ³	S	1

Preferred compounds of the formula XVII are listed in Table 17 below as compounds of the formula XVIIb:

Table 17: Compounds of the formula XVIIb

$$R_{82}$$
 Z_4 O (XVIIb)

Comp. No.	U	R ₈₂	Z_4
17.001	0	Н	C=CH COOCH3
17.002	0	Н	C=CH C=CH ₂ CH ₂
17.003	0	5-CI	C=CH COOCH3
17.004	CH₂	Н	C=CH COOCH3
17.005	CH ₂	Н	C=CH_CCH ₂ COO-CH ₂
17.006	CH ₂	Н	C=CH COOC ₂ H ₅
17.007	NH	5-CI	C=CH CH ³ CH ³
17.008	NH	5-CI	C=CH COOCH3
17.009	NH	Н	C=CH COOCH3
17.010	NH	Н	C=CH CH³ CH³

Comp.	U	R_{82}	Z_4
No.			
17.011	NCH₃	Н	C=CH CH3
17.012	NCH ₃	Н	C=CH COOCH3

Preferred compounds of the formula XVII are listed in Table 18 below as compounds of the formula XVIIc:

Table 18: Compounds of the formula XVIIc

Comp. U V r W₁ Z₄ R₈₂ No. 18.001 O C=O 1
$$C=CH_{O} CH_{2} CH_{3} CH_{2} CH_{2}$$

Preferred compounds of the formula XVII are listed in Table 19 below as compounds of the formula XVIId:

Table 19: Compounds of the formula XVIId

Comp.
$$R_{82}$$
 W_1 (XVIId)

No. 19.001 6-Cl $C = CH_{O} CH_{2}^{COOCH_{3}}$

19.002 6-Cl $C = CH_{O} CH_{2}^{CH}$

19.003 H $C = CH_{O} CH_{2}^{CH}$

19.004 H $C = CH_{O} CH_{2}^{CH}$

19.005 H $C = CH_{O} CH_{2}^{CH}$

Preferred compounds of the formula XVIII are listed in Table 20 below:

Table 20: Compounds of the formula XVIII

Comp. No.	Comp. No. R ₁₀₃		R ₁₀₅	R ₁₀₆
20.01	CH ₃	Н	cyclopropyl	Н
20.02	CH₃	C_2H_5	cyclopropyl	Н

20.03	CH ₃	cyclopropyl	C_2H_5	Н
20.04	CH ₃	CH ₃	Н	Н
20.05	CH₃	CH ₃	cyclopropyl	Н
20.06	CH₃	OCH ₃	OCH ₃	Н
20.07	CH ₃	CH ₃	OCH ₃	Н
20.08	CH ₃	OCH ₃	CH ₃	Н
20.09	CH₃	CH ₃	CH ₃	Н
20.10	C ₂ H ₅	CH₃	CH ₃	Н
20.11	C₂H₅	OCH ₃	OCH ₃	Н
20.12	Н	OCH ₃	OCH ₃	Н
20.13	Н	CH ₃	CH ₃	Н
20.14	C ₂ H ₅	Н	Н	СН3
20.15	Н	Н	Н	СН3
20.16	CH ₃	Н	Н	СН3
20.17	CH ₃	CH ₃	Н	CH ₃

The invention also relates to a method for the selective control of weeds in crops of useful plants which comprises treating the useful plants, their seeds or seedlings or the area on which they are cultivated jointly or separately with a herbicidally effective amount of the herbicide of the formula I and a herbicide-antagonistically effective amount of the safener of the formula X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII.

Crop plants which can be protected against the damaging effect of the abovementioned herbicides by the safeners of the formula X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII are, in particular, cereals, cotton, soya, sugarbeet, sugarcane, plantings, rapeseed, maize and rice, very particularly maize and cereals. Crops are to be understood as including those which have been rendered tolerant towards herbicides or classes of herbicides by conventional breeding methods or genetical engineering methods.

The weeds to be controlled can be both monocotyledonous and dicotyledonous weeds, for example the monocotyledonous weeds Avena, Agrostis, Phalaris, Lolium, Bromus, Alopecurus, Setaria, Digitaria Brachiaria, Echinochloa, Panicum, Sorghum hal./bic., Rottboellia, Cyperus, Brachiaria, Scirpus, Monochoria, Sagittaria, and Stellaria and the dicotyledonous weeds Sinapis, Chenopodium, Galium, Viola, Veronica, Matricaria, Papaver, Solanum, Abutilon, Sida, Xanthium, Amaranthus, Ipomoea and Chrysanthemum.

Areas under cultivation are the areas on which the crop plants are already growing, or on which the seeds of these crop plants have been sown, and also the soils which are intended to be cultivated with these crop plants.

Depending on the intended use, a safener of the formula X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII can be employed for pretreating the seeds of the crop plant (dressing of the seeds or the seedlings), or it can be worked into the soil before or after seeding. However, it can also be applied on its own or together with the herbicide after the plants have emerged. Thus, the treatment of the plants or the seeds with the safener can, in principle, be carried out independently of when the herbicide is applied. However, the plant can also be treated by simultaneous application of herbicide and safener (for example as tank mix). The application rate of safener to herbicide to be applied depends essentially on the type of application. In a field treatment which is carried out either by using a tank mix of a combination of safener and herbicide or by separate application of safener and herbicide, the ratio of herbicide to safener is as a rule from 100:1 to 1:10, preferably from 20:1 to 1:1. As a rule, 0.001 to 1.0 kg of safener/ha, preferably 0.001 to 0.25 kg of safener/ha are applied in the field treatment.

The application rates of herbicide are as a rule between 0.001 and 2 kg/ha, but preferably between 0.005 to 0.5 kg/ha.

The compositions according to the invention are suitable for all application methods which are customary in agriculture, for example pre-emergence application, postemergence application and seed dressing.

For seed dressing, generally 0.001 to 10 g of safener/kg of seed, preferably 0.05 to 2 g of safener/kg of seed, are applied. If the safener is applied in liquid form while swelling the seeds shortly before seeding, it is advantageous to employ safener solutions which comprise the active compound in a concentration of from 1 to 10000, preferably from 100 to 1000, ppm.

For application, the safeners of the formula X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII or combinations of these safeners with the herbicides of the formula I are advantageously processed together with auxiliaries conventionally used in the art of formulation, for example to give emulsion concentrates, spreadable pastes, directly sprayable or dilutable

solutions, dilute emulsions, wettable powders, soluble powders, dusts, granules or microcapsules.

Such formulations are described, for example, in WO 97/34485 on pages 9 to 13. The formulations are prepared in a known manner, for example by intimate mixing and/or grinding of the active compounds with liquid or solid formulation auxiliaries, for example solvents or solid carriers. Surface-active compounds (surfactants) can furthermore additionally be used during preparation of the formulations. Solvents and solid carriers which are suitable for this purpose are mentioned, for example, in WO 97/34485 on page 6.

Suitable surface-active compounds are, depending on the nature of the active compound of the formula I to be formulated, nonionic, cationic and/or anionic surfactants and surfactant mixtures having good emulsifying, dispersing and wetting properties. Examples of suitable anionic, nonionic and cationic surfactants are listed, for example, in WO 97/34485 on pages 7 and 8. The surfactants conventionally used in the art of formulation and which can also be used in the preparation of the herbicidal compositions according to the invention are described, inter alia, in "Mc Cutcheon's Detergents and Emulsifiers Annual", MC Publishing Corp., Ridgewood New Jersey, 1981, Stache, H., "Tensid-Taschenbuch" [Surfactant handbook], Carl Hanser Verlag, Munich/Vienna, 1981 and M. and J. Ash, "Encyclopedia of Surfactants", Vol I-III, Chemical Publishing Co., New York, 1980-81.

The herbicidal formulations as a rule comprise 0.1 to 99% by weight, in particular 0.1 to 95% by weight, of the active compound mixture of the compound of the formula I with the compounds of the formulae X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII, 1 to 99.9% by weight of a solid or liquid formulation auxiliary and 0 to 25% by weight, in particular 0.1 to 25% by weight, of a surfactant. While concentrated compositions are usually preferred as commercial goods, the end user as a rule uses dilute compositions.

The compositions can also comprise further additives, such as stabilizers, for example epoxidized or non-epoxidized vegetable oils (epoxidized coconut oil, rapeseed oil or soya oil), defoamers, for example silicone oil, preservatives, viscosity regulators, binders, tackifiers and fertilizers or other active substances. For using safeners of the formula X, XI, XIII, XIV, XV, XVI, XVII or XVIII or compositions comprising them to protect crop plants against damaging effects of herbicides of the formula I, various methods and techniques are suitable, for example the following:

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i) Seed dressing

- a) Dressing the seeds with an active compound of the formula X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII formulated as a wettable powder by shaking in a vessel until even distribution on the surface of the seeds is achieved (dry dressing). Here, approximately 1 to 500 g of active compound of the formula X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII (4 g to 2 kg of wettable powder) are employed per 100 kg of seed.
- b) Dressing the seeds using an emulsion concentrate of the active compound of the formula X, XI, XII, XIV, XV, XVI, XVII or XVIII according to method a) (wet dressing).
- c) Dressing by dipping the seeds for 1 to 72 hours into a liquor containing 1-1000 ppm of active compound of the formula X, XI, XIII, XIV, XV, XVI, XVII or XVIII, with or without subsequent drying of the seeds (dip dressing).

Seed dressing or the treatment of the germinated seedling are the naturally preferred application methods, since the treatment with active compound is completely directed at the target culture. As a rule, 1 to 1000 g of antidote, preferably 5 to 250 g of antidote, are employed per 100 kg of seed, but, depending on the method, which also permits the addition of other active compounds or micronutrients, it is possible to deviate above or below the stated limit concentrations (repeat dressing).

ii) Application as tank mix

A liquid preparation of a mixture of antidote and herbicide (mutual ratio between 10:1 and 1:100) is employed, the application rate of herbicide being from 0.005 to 5.0 kg per hectare. Such tank mixes are applied before or after seeding.

iii) Application in the seed farrow

The active compound of the formula X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII is applied into the open seeded seed farrow as an emulsion concentrate, a wettable powder or as granules. After the seed farrow has been covered, the herbicide is applied in a customary manner by the pre-emergence method.

iv) Controlled release of active compound

The active compound of the formula X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII is absorbed in solution onto mineral granule carriers or polymerized granules (urea/formaldehyde) and dried. If appropriate, a coating which allows the active compound to be released in metered form over a certain period of time can be applied (coated granules).

The efficacy of herbicidal and plant-growth-inhibiting compositions according to the invention comprising a herbicidally effective amount of a compound of the formula I and a herbicide-antagonistically effective amount of a compound of the formula X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII can be increased by addition of spray tank adjuvants. These adjuvants may be, for example, nonionic surfactants, mixtures of nonionic surfactants, mixtures of anionic surfactants with nonionic surfactants, cationic surfactants, organosilicon surfactants, mineral oil derivatives with or without surfactants, vegetable oil derivatives with or without addition of surfactants, alkylated derivatives of oils of vegetable or mineral origin with or without surfactants, fish oils and other oils of animal nature and their alkyl derivatives with or without surfactants, natural higher fatty acids, preferably having 8 to 28 carbon atoms, and their alkyl ester derivatives, organic acids which contain an aromatic ring system and one or more carboxylic esters, and their alkyl derivatives, furthermore suspensions of polymers of vinyl acetate or copolymers of vinyl acetate/acrylic esters. Mixtures of individual adjuvants with one another and in combination with organic solvents may further increase the effect.

Suitable nonionic surfactants are, for example, polyglycol ether derivatives of aliphatic or cycloaliphatic alcohols, saturated or unsaturated fatty acids and alkylphenols, preferably those which may contain 3 to 30 glycol ether groups and 8 to 20 carbon atoms in the (aliphatic) hydrocarbon radical and 6 to 18 carbon atoms in the alkyl radical of the alkylphenols.

Other suitable nonionic surfactants are the water-soluble polyethylene oxide adducts on polypropylene glycol, ethylenediaminopolypropylene glycol and alkylpolypropylene glycol preferably having 1 to 10 carbon atoms in the alkyl chain which preferably contain 20 to 250 ethylene glycol ether groups and 10 to 100 propylene glycol ether groups. The abovementioned compounds generally contain 1 to 5 ethylene glycol units per propylene glycol unit.

Other examples of nonionic surfactants which may be mentioned are nonylphenolpolyethoxyethanols, castor oil polyglycol ethers, polypropylene/polyethylene oxide adducts, tributylphenoxypolyethoxyethanol, polyethylene glycol and octylphenoxypolyethoxyethanol.

Also suitable are fatty esters of polyoxyethylene sorbitan, for example polyoxyethylene sorbitan trioleate.

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Preferred anionic surfactants are, in particular, alkyl sulfates, alkyl sulfonates, alkylaryl sulfonates, alkylated phosphoric acids and their ethoxylated derivatives. The alkyl radicals usually contain 8 to 24 carbon atoms.

Preferred nonionic surfactants are known under the following trade names:

Polyoxyethylene cocoalkylamine (for example AMIET® 105 (Kao Co.)), polyoxyethylene oleylamine (for example AMIET® 415 (Kao Co.)), nonylphenolpolyethoxyethanols, polyoxyethylene stearylamine (for example AMIET® 320 (Kao Co.)), N-polyethoxyethylamines (for example GENAMIN® (Hoechst AG)), N,N,N',N'-tetra(polyethoxypolypropoxyethyl)ethylene diamines (for example TERRONIL® and TETRONIC® (BASF Wyandotte Corp.)), BRIJ® (Atlas Chemicals), ETHYLAN® CD and ETHYLAN® D (Diamond Shamrock), GENAPOL® C, GENAPOL® O, GENAPOL® S and GENAPOL® X080 (Hoechst AG), EMULGEN® 104P, EMULGEN® 109P and EMULGEN® 408 (Kao Co.); DISTY® 125 (Geronazzo), SOPROPHOR® CY 18 (Rhône Poulenc S.A.); NONISOL® (Ciba-Geigy), MRYJ® (ICI); TWEEN® (ICI); EMULSOGEN® (Hoechst AG); AMIDOX® (Stephan Chemical Co.), ETHOMID® (Armak Co.); PLURONIC® (BASF Wyandotte Corp.), SOPROPHOR® 461P (Rhône Poulenc S.A.), SOPROPHOR® 496/P (Rhône Poulenc S.A.), ANTAROX FM-63 (Rhône Poulenc S.A.), SLYGARD 309 (Dow Corning), SILWET 408, SILWET L-7607N (Osi-Specialities).

The cationic surfactants are primarily quaternary ammonium salts which contain, as N-substituents, at least one alkyl radical having 8 to 22 C atoms and, as further substituents, lower nonhalogenated or halogenated alkyl, benzyl or lower hydroxyalkyl radicals. The salts are preferably present as halides, methyl sulfates or ethyl sulfates, for example stearyltrimethylammonium chloride or benzyldi(2-chloroethyl)ethylammonium bromide.

The oils used are either of mineral or natural origin. The natural oils may additionally be of animal or vegetable origin. In the case of animal oils, preference is given, in particular, to derivatives of beef tallow, but fish oils (for example sardine oil) and derivatives thereof are also used. Vegetable oils are mainly seed oils of various origin. Examples of particularly preferred vegetable oils which may be mentioned are coconut, rapeseed or sunflower oils and derivatives thereof.

Surfactants, oils, in particlar vegetable oils, derivatives thereof such as alkylated fatty acids and mixtures thereof, for example with preferably anionic surfactants such as alkylated phosphoric acids, alkyl sulfates and alkylaryl sulfonates and higher fatty acids which are customary in formulation and adjuvant technique and which can also be employed in the compositions according to the invention and spray tank solutions thereof are described, inter alia, in "Mc Cutcheon's Detergents and Emulsifiers Annual", MC Publishing Corp., Ridgewood New Jersey, 1998, Stache, H., "Tensid-Taschenbuch" [Surfactant handbook], Carl Hanser Verlag, Munich/Vienna, 1990, M. and J. Ash, "Encyclopedia of Surfactants", Vol. I-IV, Chemical Publishing Co., New York, 1981-89, G. Kapusta, "A Compendium of Herbicide Adjuvants", Southern Illinois Univ., 1998, L. Thomson Harvey, "A Guide to Agricultural Spray Adjuvants Used in the United States", Thomson Pubns., 1992.

In particular, preferred formulations have the following compositions: (% = per cent by weight)

Emulsifiable concentrates:

active compound mixture:

1 to 90%, preferably 5 to 20%

surface-active agent:

1 to 30%, preferably 10 to 20%

liquid carrier:

5 to 94%, preferably 70 to 85%

Dusts:

active compound mixture:

0.1 to 10%, preferably 0.1 to 5%

solid carrier:

99.9 to 90%, preferably 99.9 to 99%

Suspension concentrates:

active compound mixture:

5 to 75%, preferably 10 to 50%

water:

94 to 24%, preferably 88 to 30%

surface-active agent:

1 to 40%, preferably 2 to 30%

Wettable powders:

active compound mixture: 0.5 to 90%, preferably 1 to 80%

surface-active agent:

0.5 to 20%, preferably 1 to 15%

solid carrier material:

5 to 95%, preferably 15 to 90%

Granules:

active compound mixture: 0.1 to 30%, preferably 0.1 to 15%

solid carrier:

99.5 to 70%, preferably 97 to 85%

The following examples illustrate the invention in more detail, without limiting it. Formulation examples for mixtures of herbicides of the formula I and safeners of the formula X. XI. XII. XIV. XV. XVI. XVII or XVIII (% = per cent by weight)

TOTTIGIA X, XI, XII, XIII, XIV, XV,	AVI, AVII OF	XVIII (% = per cen	t by weight)	
F1. Emulsion concentrates	a)	b)	c)	d)
Active compound mixture	5%	10%	25%	50%
Ca dodecylbenzenesulfonate	6%	8%	6%	8%
Castor oil polyglycol ether	4%	· -	4%	4%
(36 mol of EO)				
Octylphenol polyglycol ether	-	4%	-	2%
(7-8 mol of EO)				
Cyclohexanone	-	-	10%	20%
Arom. hydrocarbon	85%	78%	55%	16%
mixture C ₉ -C ₁₂				.070

Emulsions of any desired concentration can be prepared from such concentrates by dilution with water.

F2. Solutions	a)	b)	c)	d)
Active compound mixture	5%	10%	50%	90%
1-Methoxy-3-(3-methoxy-				30,0
propoxy)propane	-	20%	20%	_
Polyethylene glycol MW 400	20%	10%	-	-
N-Methyl-2-pyrrolidone	-	-	30%	10%
Arom. hydrocarbon	75%	60%	•	-
mixture C ₉ -C ₁₂				

The solutions are suitable for use in the form of tiny droplets.

F3. Wettable powders	a)	b)	c)	d)
Active compound mixture	5%	25%	50%	80%
Sodium lignosulfonate	4%	-	3%	-
Sodium laurylsulfate	2 %	3 %	-	4 %
Sodium diisobutylnaphthalene-		6%	5%	6%
sulfonate				
Octylphenol polyglycol ether	-	1%	2%	-
(7-8 mol of EO)				
Finely divided silica	1%	3%	5%	10%
Kaolin	88%	62%	35%	•

The active compound is thoroughly mixed with the additives and ground well in a suitable mill. This gives spray powders which can be diluted with water to give suspensions of any desired concentration.

F4. Coated granules	a)	b)	c)
Active compound mixture	0.1%	5%	15%
Finely divided silica	0.9%	2%	2%
Inorg. carrier material	99.0%	93%	83%
(Æ 0.1 - 1 mm),			

for example CaCO₃ or SiO₂

The active compound is dissolved in methylene chloride, the solution is sprayed onto the carrier and the solvent is subsequently evaporated off under reduced pressure.

F5. Coated granules	a)	b)	c)
Active compound mixture	0.1%	5%	15%
Polyethylene glycol MW 200	1.0%	2%	3%
Finely divided silica	0.9%	1%	2%
Inorg. carrier material	98.0%	92%	80%
(Æ 0.1 - 1 mm),			

for example CaCO₃ or SiO₂

In a mixer, the finely ground active compound is applied evenly to the carrier material moistened with polyethylene glycol. In this manner, dust-free coated granules are obtained.

F6. Extruder granules	a)	b)	c)	d)
Active compound mixture	0.1%	3%	5%	15%
Sodium lignosulfonate	1.5%	2%	3%	4%

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Carboxymethylcellulose	1.4%	2%	2%	2%
Kaolin	97.0%	93%	90%	79%

The active compound is mixed with the additives, ground and moistened with water. This mixture is extruded and subsequently dried in a stream of air.

F7. Dusts	a)	b)	c)
Active compound mixture	0.1%	1%	5%
Talc mixture	39.9%	49%	35%
Kaolin	60.0%	50%	60%

Ready-to-use dusts are obtained by mixing the active compound with the carriers and grinding the mixture in a suitable mill.

F8. Suspension concentrates	a)	b)	c)	d)
Active compound mixture	3%	10%	25%	50%
Ethylene glycol	5%	5%	5%	5%
Nonylphenol polyglycol ether	-	1%	2%	
-				
(15 mol of EO)				
Sodium lignosulfonate	3%	3%	4%	5%
Carboxymethylcellulose	1%	1%	1%	1%
37% aqueous formaldehyde	0.2%	0.2%	0.2%	0.2%
solution				
Silicone oil emulsion	0.8%	0.8%	0.8%	0.8%
Water	87%	79%	62%	38%

The finely ground active compound is intimately mixed with the additives. This gives a suspension concentrate, from which suspensions of any desired concentration can be prepared by dilution with water.

It is often more useful to formulate the active compound of the formula I and the mixing partner of the formula X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII individually and then to mix them shortly before application in the applicator in the desired mixing ratio as "tank mix" in water.

The capability of the safeners of the formula X, XI, XII, XIII, XIV, XV, XVI, XVII or XVIII to protect crop plants against the phytotoxic action of herbicides of the formula I is illustrated in the examples below.

Biological Example 1: Safening action

Under greenhouse conditions, the test plants are grown in plastic pots until they have reached the 4-leaf-stage. In this stage, both the herbicide on its own and the mixtures of the herbicide with the test substances to be tested as safeners are applied to the test plants. The application is carried out as an aqueous suspension of the test substances, prepared from a 25% wettable powder (Example F3, b)), using 500 l of water/ha. 3 weeks after the application, the phytotoxic effect of the herbicide on the crop plants, for example maize and cereals, is evaluated using a percentage scale. 100% means that the test plant has died, 0% means no phytotoxic effect.

The results obtained in this test show that the damage to the crop plants caused by the herbicide of the formula I can be considerably reduced using the compounds of the formula X, XI, XII, XIV, XV, XVI, XVII or XVIII.

The same results are obtained when the mixtures are formulated in accordance with Examples F1, F2 and F4 to F8.

Biological Example 2: Safening of the compound no. 1.032

Under greenhouse conditions, the test plants are grown in plastic pots until they have reached the 4-leaf-stage. In this stage, both the herbicide on its own and the mixtures of the herbicide with the test substances to be tested as safeners are applied to the test plants. The application is carried out as an aqueous suspension of the test substances, prepared from an emulsion concentrate (EC 100; Example F1) of the herbicides and an emulsion concentrate (EC 100; Example F1) of the safeners (exceptions: the safeners no. 10.05 and 20.17, which are employed as a 25% wettable powder (Example F3, b)). 9 days after the application, the phytotoxic effect of the herbicide on summer wheat and durum wheat is evaluated using a percentage scale (100%: test plant has died; 0%: no phytotoxic effect).

Table S2: Safening of the compound no. 1.032

Herbicide	1.032	1.032	1.032	1.032
no.		+	+	+
+		10.01	9.01	10.05
safener no.				
Application	250 125 60	250 125 60	250 125 60	250 125 60
rate (g/ha)	+ + +	+ + +	+ + +	+ + +
	0 0 0	60 30 15	60 30 15	60 30 15
Summer	30 20 10	0 0 0	5 0 0	0 0 0
wheat				
Durum	20 5 0	10 5 0	0 0 0	000
wheat				

Herbicide	1.032	1.032	1.032	1.032
no.		+	+	+
+		20.17	9.02	11.03
safener no.				
Application	250 125 60	250 125 60	250 125 60	250 125 60
rate (g/ha)	+ + +	+ + +	+ + +	+ + +
	0 0 0	60 30 15	60 30 15	60 30 15
Summer	30 20 10	10 5 0	0 0 0	0 0 0
wheat		_		
Durum	20 5 0	0 0 0	0 0 0	0 0 0
wheat				

Biological Example 3: Safening of the compound no. 1.025

Under greenhouse conditions, the test plants are grown in plastic pots until they have reached the 4-leaf-stage. In this stage, both the herbicide on its own and the mixtures of the herbicide with the test substances to be tested as safeners are applied to the test plants. The application is carried out as an aqueous suspension of the test substances, prepared from an emulsion concentrate (EC 100; Example F1) of the herbicides and an emulsion concentrate (EC 100; Example F1) of the safeners (exceptions: the safeners no. 10.05 and 20.17, which are employed as a 25% wettable powder (Example F3, b)). 11 days after the application, the phytotoxic effect of the herbicide on summer wheat and durum wheat is evaluated using a percentage scale (100%: test plant has died; 0%: no phytotoxic effect).

Table S3: Safening of the compound no. 1.025

Herbicide	1.025	1.025	1.025	1.025
no.		+	+	+
+		10.01	9.01	10.05
safener no.				
Application	500 250 125	500 250 125	500 250 125	500 250 125
rate (g/ha)	+ + +	+ + +	+ + +	+ + +
	0 0 0	125 60 30	125 60 30	125 60 30
Summer wheat	55 40 10	10 0 0	0 0 0	5 0 0
Durum wheat	40 5 0	10 0 0	0 0 0	0 0 0

Herbicide	1.025	1.025	1.025	1.025
no.		+	+	+
+		20.17	9.02	11.03
safener no.				
Application	500 250 125	500 250 125	500 250 125	500 250 125
rate (g/ha)	+ + +	+ + +	+ + +	+ + +
	0 0 0	125 60 30	125 60 30	125 60 30
Summer wheat	55 40 10	10 5 5	20 5 0	10 5 0
Durum wheat	40 5 0	0 0 0	5 0 0	0 0 0

Biological Example 4: Safening of the compound no. 1.007

Under greenhouse conditions, the test plants are grown in plastic pots until they have reached the 4-leaf-stage. In this stage, both the herbicide on its own and the mixtures of the herbicide with the test substances to be tested as safeners are applied to the test plants. The application is carried out as an aqueous suspension of the test substances, prepared from an emulsion concentrate (EC 100; Example F1) of the herbicides and an emulsion concentrate (EC 100; Example F1) of the safeners (exceptions: the safeners no. 10.05 and 20.17, which are employed as a 25% wettable powder (Example F3, b)). 9 days after the application, the phytotoxic effect of the herbicide on summer wheat and durum wheat is evaluated using a percentage scale (100%: test plant has died; 0%: no phytotoxic effect).

Table S4: Safening of the compound no. 1.007

Herbicide	1.007	1.007	1.007	1.007
no.		+	+	+
+		10.01	9.01	10.05
safener no.				
Application	250 125 60	250 125 60	250 125 60	250 125 60
rate (g/ha)	+ + +	+ + +	+ + +	+ + +
	0 0 0	60 30 15	60 30 15	60 30 15
Summer wheat	60 60 60	30 20 10	20 10 0	30 20 10
Durum wheat	60 60 55	20 10 5	10 5 0	20 10 5

Herbicide	1.007	1.007	1.007	1.007
no.		+	+	+
+		20.17	9.02	11.03
safener no.				
Application	250 125 60	250 125 60	250 125 60	250 125 60
rate (g/ha)	+ + +	+ + +	+ + +	+ + +
	0 0 0	60 30 15	60 30 15	60 30 15
Summer	60 60 60	60 60 40	20 10 10	20 10 10
wheat				
Durum	60 60 55	60 50 40	10 5 5	10 5 5
wheat				

The following examples illustrate the invention in more detail without limiting it.

Preparation Examples:

Example H1: Preparation of:

Over a period of one hour, a solution of 177.6 g of methanesulfonyl chloride and 400 ml of diethyl ether is added dropwise to a solution of 80.6 g (0.76 mol) of diethylene glycol and 159.9 g (1.58 mol) of triethylamine in 1500 ml of diethyl ether which had been cooled to -10°C, and during the addition, the temperature is kept below 5°C. The mixture is stirred at a temperature of 0°C for 30 minutes, and cooling is then removed. After 2 hours, at a temperature of 20°C, 12 ml of triethylamine and 12 ml of methanesulfonyl chloride are added, and stirring is continued for another 4 hours. The resulting white suspension is subsequently transferred onto a suction filter, and the residue is washed twice with 300 ml of diethyl ether. The filter cake is taken up in 2000 ml of ethyl acetate, and the suspension is stirred at room temperature for 30 minutes and then filtered again. The resulting filtrate is concentrated and the residue is used for the next reaction without any further purification. 216.5 g of the desired crude product (1) are obtained in the form of white crystals.

Example H2:

$$CH_{3}SO_{2}O$$

$$+ H_{3}C$$

$$+ H_$$

A solution of 68.78 g (0.30 mol) of (2) in 140 ml of dimethylformamide is added dropwise for a period of 30 minutes to a suspension of 23.9 g (0.60 mol) of 60% sodium hydride in 500 ml of dimethylformamide which had been cooled to 5°C. Cooling is removed and the reaction mixture is stirred until it has reached a temperature of 20°C. The mixture is subsequently briefly heated to a temperature of from 30 to 40°C to bring the evolution of hydrogen to completion. After cooling to a temperature of from 0 to 5°C, a solution of 80 g (0.305 mol) of (1) in 160 ml of dimethylformamide is added dropwise over a period of

30 minutes, during which the temperature is kept at from 0 to 5°C. Cooling is removed and the reaction mixture is stirred at room temperature for 3 hours and at approximately 40°C for 45 minutes and then added to a mixture of saturated ammonium chloride solution, ice and tert-butyl methyl ether. The phases are separated and the organic phase is subsequently washed with water (2x). The organic phase is dried with sodium sulfate and evaporated, and the residue is dried further at a temperature of 40°C and under reduced pressure, giving 92.2 g of (3) in the form of a slightly yellow oil. The crude product is employed for the next reaction without any further purification.

Example H3:

160.5 ml of a 33% solution of hydrogen bromide in glacial acetic acid are added dropwise over a period of 30 minutes to a solution of 92.2 g (0.305 mol) of (3) in 1200 ml of diethyl ether which had been cooled to 0°C. Cooling is removed and the mixture is subsequently stirred at 20°C for 22 hours and then under reflux for 27 hours, the resulting white suspension is transferred onto a suction filter and washed with diethyl ether, and the filter residue is subsequently dried over P_2O_5 under reduced pressure at a temperature of from 50 to 60°C. The product (4) is obtained in a yield of 52.9 g in the form of a white solid.

Example H4:

71.8 g (0.71 mol) of triethylamine are added to a suspension of 40 g (0.15 mol) of (4) in 1000 ml of xylene, and the mixture is degassed (4 x vacuum/argon). The yellow suspension

is subsequently heated to a temperature of 60°C and stirred for 3 hours. 42.5 g (0.15 mol) of (5) are then added, and the mixture is heated to a bath temperature of 150°C to distil off excess triethylamine and the ethanol which is formed. After 3 hours, the reaction mixture is cooled to a temperature of 40°C and added to 500 ml of an ice/water mixture. Using 100 ml of aqueous 1N sodium hydroxide solution, the reaction mixture is made alkaline and the aqueous phase (which contains the product) is washed twice with ethyl acetate. The organic phase is reextracted twice using aqueous 1N sodium hydroxide solution, the aqueous phases are combined, the remaining xylene is distilled off and the combined aquoeus phases are adjusted to pH 2-3 using 4N HCl with cooling. The product which precipitates is transferred onto a suction filter, the filter cake is washed with water and briefly with hexane and is subsequently dried under reduced pressure at a temperature of 60°C over P₂O₅. This gives 34.6 g of (6) as a slightly beige solid of melting point 242-244°C (decomp.).

Example H5:

$$H_3C$$
 CH_3
 CH_3

A catalytic amount of 4-dimethylaminopyridine is added to a solution of 3 g (10.4 mmol) of (6) and 1.6 g (15.8 mmol) of triethylamine in 100 ml of tetrahydrofuran which had been cooled to a temperature of 0°C. 1.57 g (13.0 mmol) of pivaloyl chloride are subsequently added dropwise. The mixture is stirred at a temperature of 0°C for 30 minutes, cooling is removed, and the mixture is stirred for a further 60 minutes. The reaction mixture is subsequently poured into saturated aqueous sodium chloride solution, and the organic phase is separated off. The organic phase is dried over magnesium sulfate, filtered off and concentrated. Chromatographic purification and recrystallization from diethyl ether gives 2.94 g of (7) of melting point 135-136°C.

Example H6: Preparation of

$$H_2C$$
OMe
 H_3C
OMe
 H_2C
OMe

First 36.7 g (0.116 mol) of tributylvinylstannane and then 2 g of tetrakis(triphenylphosphine)palladium are added to a solution of 20 g of dimethyl 2-(2,6-dibromo-4-methyl-phenyl)malonate (52.6 mmol) in 400 ml of toluene (3 x degassed, vacuum/argon). The reaction mixture is then stirred at a temperature of from 90 to 95°C for 9 hours. Filtration through Hyflo and concentration on a rotary evaporator gives, after chromatographic purification, 15.3 g of (8) in the form of a yellow oil which is used for the next reaction without any further purification.

Example H7:

$$H_3C$$
OMe
 H_2
OMe
 H_3C
OMe

At a temperature of from 20 to 25°C, 15.2 g of the compound (8) obtained according to Example H6 are hydrogenated with hydrogen over a palladium catalyst (using carbon as carrier, 7 g of 5% Pd/C) in 160 ml of tetrahydrofuran. After the hydrogenation has ended, the product is filtered through Hyflo, and the resulting filtrate is concentrated on a rotary evaporator. This gives 13.7 g of (9) in the form of yellow crystals of melting point 47 - 49°C.

Example H8:

By the method of Preparation Example H4, but starting from 4.8 g (17.2 mmol) of the malonate (9), 4.56 g of the compound (10) are obtained as a solid of melting point 188-190°C.

Example H9:

A catalytic amount of 4-dimethylaminopyridine is added to a solution of 1 g (3.2 mmol) of (10) and 0.65 g (6.4 mmol) of triethylamine in 30 ml of tetrahydrofuran which had been cooled to a temperature of 0°C. 0.49 g (4.1 mmol) of pivaloyl chloride is subsequently added dropwise. The mixture is stirred at a temperature of 0°C for 10 minutes, cooling is removed, and stirring is then continued for a further 90 minutes. The reaction mixture is poured into saturated aqueous sodium chloride solution and diluted with tert-butyl methyl ether, and the organic phase is separated off. The organic phase is dried over magnesium sulfate, filtered off and concentrated. Chromatographic purification gives 1.07 g of (11) in the form of a white solid of melting point 122-123°C.

Example H10:

67.8 g (0.59 mol) of methanesulfonyl chloride are added dropwise to a solution of 37.1 g (0.28 mol) of *cis*-2,5-bis(hydroxymethyl)tetrahydrofuran (12) and 65.3 g (0.65 mol) of triethylamine in 400 ml of methylene chloride which had been cooled to 0-3°C, during which the temperature is kept below 7°C. The mixture is subsequently stirred at a temperature of 20°C overnight. The resulting white suspension is transferred onto a suction filter, the residue is washed with methylene chloride and the filtrate is concentrated. The residue is taken up in ethyl acetate, washed with water (2x) and with saturated aqueous sodium chloride solution (1x), dried (Na₂SO₄) and concentrated. This gives 72.7 g of the dimesylate compound (13) as a crude oil which is employed for the next reaction without any further purification.

The starting material (12) is known from the literature: see, for example, K. Naemura et al., Tetrahedron Asymmetry 1993, 4, 911-918.

(14)

Example H11:

(13)

By the method of Preparation Example H2, but starting from 21.0 g (0.53 mol) of 60% NaH, 58.4 g (0.25 mol) of (2) and 72.5 g (0.25 mol) of dimesylate (13) in a total of 840 ml of dimethylformamide, (14) is obtained as a crude brown oil. Chromatographic purification gives 53.7 g of the pure compound (14) as a white solid of melting point 81 - 83°C.

$$H_3C$$
 H_3C
 H_3C

Example H12:

By the method of Preparation Example H3, but starting from 53.5 g (0.16 mol) of (14) in 800 ml of diethyl ether and 90 ml of a 33% solution of hydrogen bromide in conc. acetic acid, 36.5 g of the bicyclic hydrazine (15) are obtained as a solid of melting point 262 - 264°C.

Example H13:

By the method of Preparation Example H4, but starting from 0.105 mol of the malonate (9) and 30.4 g (0.105 mol) of the hydrazine (15), 29.7 g of the compound (16) are obtained as a solid of melting point 287°C.

Example

<u>H14:</u>

$$H_3C$$
 H_3C
 H_3C

By the method of Preparation Example H9, but starting from 1.1 g (3.2 mmol) of (16), 0.83 g of the pivaloyl ester (17) is obtained as a solid of melting point 141-143°C.

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Table 1: Compounds of the formula le:

$$R_2 \xrightarrow{R_1} 0 \\ N \\ N \\ O$$
 (le)

Comp. No.	R ₁	R_2	R ₃	G	Phys. data
1.001	CH ₃	СН₃	CH ₃	Н	m.p. 245°C
1.002	CH₃	CH ₃	CH ₃	C(O)C(CH ₃) ₃	m.p. 135- 136°C
1.003	CH₃	CH ₃	CH ₃	C(O)OCH₂CH₃	
1.004	CH₂CH₃	CH₃	CH₃	Н	m.p. 182- 185°C
1.005	CH₂CH₃	CH₃	CH ₃	C(O)C(CH ₃) ₃	m.p. 110- 113°C
1.006	CH₂CH₃	CH₃	СН₃	C(O)OCH₂CH₃	
1.007	CH₂CH₃	CH₃	CH ₂ CH ₃	Н	m.p. 189- 191°C
1.008	CH₂CH₃	CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃	m.p. 122- 124°C
1.009	CH₂CH₃	CH₃	CH₂CH₃	C(O)OCH ₂ CH ₃	m.p. 114- 116°C
1.010	CH=CH ₂	CH ₃	CH ₃	н	m.p. 165- 170°C
1.011	CH=CH ₂	CH₃	CH₃	C(O)C(CH ₃) ₃	m.p. 111- 113°C
1.012	CH=CH₂	СН₃	CH₂CH₃	Н	
1.013	CH=CH₂	CH ₃	CH=CH₂	Н	
1.014	CH=CH ₂	CH ₃	CH=CH ₂	C(O)C(CH ₃) ₃	

Comp. No.	R ₁	R ₂	R_3	G	Phys. data
1.015	C≡CH	CH₃	CH ₃	Н	m.p. 179- 184°C
1.016	C≡CH	CH ₃	CH ₃	C(O)C(CH ₃) ₃	m.p. 109- 111°C
1.017	C≡CH	CH ₃	CH ₃	C(O)OCH₂CH₃	
1.018	C≡CH	CH₃	CH₂CH₃	Н	m.p. 189- 193°C
1.019	C≡CH	CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃	
1.020	C≡CH	CH ₃	CH ₂ CH ₃	C(O)OCH ₂ CH ₃	
1.021	C≡CH	CH ₃	C≡CH	Н	m.p. 300°C
1.022	C≡CH	CH₃	C≡CH	C(O)C(CH ₃) ₃	m.p. 183- 185°C
1.023	C≅CH	CH ₃	C≡CH	C(O)OCH ₂ CH ₃	
1.024	C≡CH	CH₃	CH=CH ₂	Н	
1.025	C≡CCH ₃	CH₃	CH₃	Н	m.p. 179- 181°C
1.026	C≡CCH ₃	CH₃	CH₃	C(O)C(CH ₃) ₃	m.p. 128- 129°C
1.027	C≡CCH ₃	CH ₃	CH ₃	C(O)OCH ₂ CH ₃	
1.028	C≡CCH ₃	CH ₃	CH₂CH₃	Н	
1.029	C≡CCH ₃	CH ₃	CH ₂ CH ₃	C(O)C(CH ₃) ₃	
1.030	C≡CCH ₃	CH ₃	C≡CCH ₃	Н	
1.031	C≡CCH ₃	CH ₃	C≡CCH ₃	C(O)C(CH ₃) ₃	
1.032	CH₂CH₂CH₃	CH₃	CH₃	Н	m.p. 136- 138°C
1.033	CH₂CH₂CH₃	CH ₃	CH ₃	C(O)C(CH ₃) ₃	m.p. 65- 67°C
1.034	CH₂CH₂CH₃	CH₃	CH ₃	C(O)OCH₂CH₃	
1.035	CH₂CH₂CH₃	CH ₃	CH₂CH₃	Н	
1.036	CH₂CH₂CH₃	CH ₃	CH₂CH₂CH₃	Н	

Comp.	R ₁	R ₂	R ₃	G	Phys. data
No. 1.037	CH ₂ CH ₂ CH ₃	CH₃		C(O)C(CH ₃) ₃	
1.038	CH ₂ CH ₂ CH ₃	CH ₃	CH ₂ CH ₂ CH ₃	C(O)OCH ₂ CH ₃	
1.039	CH₂CH₂CH₃	СН₃	C≝CH	Н	
1.040	CH(CH ₃) ₂	CH₃	CH ₃	Н	m.p. 214-
1.0 10					216°C
1.041	CH(CH ₃) ₂	CH₃	CH₃	C(O)C(CH ₃) ₃	m.p. 148-
					151°C
1.042	CH(CH ₃) ₂	CH₃	CH₂CH₃	Н	
1.043	CH(CH ₃) ₂	CH₃	C≡CH	Н	
1.044	\triangleright	CH ₃	CH ₃	н	
1.045	<u> </u>	CH₃	CH₂CH₃	Н	
1.040					
1.046		CH₃	C≡CH	Н	
1.047	CH ₂ CH=CH ₂	CH₃	CH₃	Н	
1.048	CH ₂ CH=CH ₂	CH₃	CH₂CH₃	Н	
1.049	CH ₂ CH=CH ₂	CH₃	C≡CH	Н	
1.050	CH ₂ CH ₂ CH ₂ CH ₃	CH₃	CH ₃	Н	
1.051	CH ₂ CH ₂ CH ₂ CH ₃	CH₃	CH₂CH₃	Н	
1.052	N(CH ₂ CH ₃) ₂	CH₃	CH ₃	Н	
1.053	N(CH ₂ CH ₃) ₂	CH₃	CH₂CH₃	Н	
1.054	CH₂OH	CH₃	CH ₃	Н	
1.055	CH ₂ OCH ₃	CH ₃	CH ₃	Н	
1.056	CH ₂ OC(CH ₃) ₃	CH₃	CH₃	Н	
1.057	CH₃	CH₂CH₃	CH ₃	Н	
1.058	CH₂CH₃	CH₂CH₃	CH ₃	Н	
1.059	CH₂CH₃	CH₂CH₃	CH₂CH₃	Н	m.p. 185-
					187°C
1.060	CH₂CH₃	CH₂CH₃	CH₂CH₃	C(O)C(CH ₃) ₃	m.p. 126-
					128°C
1.061	CH₂CH₃	CH₂CH₃	CH ₂ CH ₃	C(O)OCH ₂ CH ₃	m.p. 105-

Comp.	R ₁	R_2	R ₃	G	Phys. data
140.					107°C
1.062	CH=CH ₂	CH₂CH₃	CH=CH ₂	Н	
1.063	C≡CH	CH₂CH₃	C≡CH	Н	
1.064	CH₃	CH=CH ₂	CH₃	Н	
1.065	CH₂CH₃	CH=CH₂	CH₂CH₃	Н	
1.066	CH₂CH₃	CH=CH ₂	CH₃	Н	
1.067	CH₂CH₃	CH=CH ₂	CH ₃	$C(O)C(CH_3)_3$	m.p. 108-
					110°C
1.068	C≡CH	CH=CH ₂	C≡CH	H	
1.069	CH₃	C≡CH	CH ₃	Н	
1.070	CH₂CH₃	C≡CH	CH ₃	Н	m.p. 240-
					243°C
1.071	CH₂CH₃	C≡CH	CH₃	C(O)C(CH ₃) ₃	m.p. 138- 140°C
1.072	CH₂CH₃	C≘CH	CH ₃	C(O)OCH₂CH₃	
1.073	CH₂CH₃	C≡CH	CH₂CH₃	Н	
1.074	CH₂CH₃	C≡CH	C≡CH	н	
1.075	C≡CH	C≡CH	C≡CH	н	
1.076	CH₃	CH ₂ CH=CH ₂	СН₃	Н	
1.077	CH ₃	CH₂CH=CH₂	CH₂CH₃	н	
1.078	CH₃	CH ₃	Br	Н	m.p. 234-
					237°C
1.079	CH₃	CH₃	Br	C(O)C(CH ₃) ₃	m.p. 76-
		-			78°C
1.080	CH ₃	CH₃	Br	C(O)OCH₂CH₃	
1.081	CH₂CH₃	CH₃	Br	Н	
1.082	C≡CH	CH ₃	Br	Н	
1.083	CH ₃	Br	CH₃	Н	m.p. 298-
		_			299°C
1.084	CH₂CH₃	Br	CH₃	Н	m.p. 261-
					263°C

Comp.	R ₁	R_2	R ₃	G	Phys. data
No. 1.085	CH₂CH₃	Br	CH₃	C(O)C(CH ₃) ₃	m.p. 127-
					130°C
1.086	CH₂CH₃	Br	CH ₃	C(O)OCH ₂ CH ₃	
1.087	CH₂CH₃	Br	CH₂CH₃	Н	
1.088	Br	CH₃	Br	Н	m.p. 238-
					241°C
1.089	Br	CH₃	Br	C(O)C(CH ₃) ₃	solid
1.090	Br	CH ₃	Br	C(O)OCH₂CH₃	
1.091	CH ₃	Br	Br	Н	
1.092	CH₂CH₃	Br	Br	Н	
1.093	CH ₃	CH ₃	Cl	Н	
1.094	CH₂CH₃	CH ₃	Cl	Н	
1.095	CH ₃	CI	CH ₃	Н	
1.096	CH₂CH₃	CI	CH ₃	Н	
1.097	CH₂CH₃	CI	CH₂CH₃	Н	
1.098	CH₂CH₃	F	CH₂CH₃	Н	
1.099	CH₂CH₃	F	C≡CH	Н	
1.100	CH₂CH₃	F	OCH₃	Н	
1.101	CI	CH ₃	Cl	Н	
1.102	CH₃	CI	Cl	Н	
1.103	CH₂CH₃	CI	Cl	Н	
1.104	Br	CH₃	Cl	Н	
1.105	CH₃	Br	CI	Н	
1.106	CH₃	CI	Br	Н	
1.107	CH₂CH₃	Br	Cl	Н	
1.108	CH₂CH₃	CI	Br	н	
1.109	OCH ₃	CH₃	CH ₃	Н	
1.110	OCH₃	CH₃	CH₂CH₃	Н	m.p. 178-
					179°C
1.111	OCH₃	CH₃	CH₂CH₃	C(O)C(CH ₃) ₃	m.p. 146-
					147°C

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Comp.	R ₁	R_2	R_3	G	Phys. data
No. 1.112	OCH ₃	CH₃	CH₂CH₃	C(O)OCH₂CH₃	
1.113	OCH ₃	CH ₃	CH ₂ CH ₂ CH ₃	Н	
1.114	OCH ₃	CH ₃	C≡CH	Н	
1.115	OCH ₃	CH₃	Br	Н	
1.116	OCH ₃	CH ₃	OCH ₃	Н	
1.117	C(O)CH ₃	CH ₃	CH ₃	Н	solid
1.118	C(O)CH ₃	CH₃	CH₂CH₃	Н	
1.119	CH ₃	C(O)CH₃	CH₂CH₃	$C(O)C(CH_3)_3$	m.p. 163-
					165°C
1.120	CH ₃	CH₂OH	CH ₂ CH ₃	Н	
1.121	CH ₃	CH ₃	CH ₃	SO ₂ CH ₂ CHCH ₂	
1.122	CH ₃	CH ₃	CH₃	SO₂CH₂CHCHCI	
1.123	CH₃	CH ₃	CH₃	SO₂CH₂CHCHCH₃	
1.124	CH₂CH₃	CH₃	CH₂CH₃	SO ₂ CH ₂ CHCH ₂	
1.125	CH₂CH₃	CH₃	CH₂CH₃	SO₂CH₂CHCHCI	
1.126	CH₂CH₃	CH ₃	CH ₂ CH ₃	SO ₂ CH ₂ CHCHCH ₃	

Table 2: Compounds of the formula If:

$$R_{2} \xrightarrow{R_{1}} O \xrightarrow{R_{21}} O \text{ (If)}$$

Comp.	R ₁	R ₂	R_3	G	R ₂₁	Phys. data
No.						-
2.001	CH ₃	CH ₃	CH ₃	Н	CH_3	
2.002	CH ₃	CH ₃	CH ₃	$C(O)C(CH_3)_3$	CH ₃	
2.003	CH ₃	CH ₃	CH ₃	C(O)OCH ₂ CH ₃	CH ₃	
2.004	CH₂CH₃	CH ₃	CH ₃	Н	CH ₃	
2.005	CH₂CH₃	CH₃	CH₂CH₃	Н	СН₃	
2.006	CH ₂ CH ₃	CH ₃	CH₂CH₃	$C(O)C(CH_3)_3$	CH ₃	

Comp.	R ₁	R_2	R ₃	G	R ₂₁	Phys. data
No.	CH₂CH₃	CH₃	CH₂CH₃	C(O)OCH₂CH₃	СНз	
2.007 2.008	CH₂CH₃	CH₃	Br	H	CH ₃	
2.008	CH₂CH₃	CH₃	Br	C(O)C(CH ₃) ₃	CH₃	
2.009	CH ₂ CH ₃	CH₃	Br	C(O)OCH ₂ CH ₃	CH ₃	
2.010	CH₂CH₃	CH₂CH₃	CH₂CH₃	H	CH ₃	
2.012	CH₂CH₃	CH₂CH₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH₃	
2.013	CH₂CH₃	CH ₂ CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH₃	
2.014	C≡CH	CH₃	CH ₃	Н	CH₃	
2.015	C≡CH	CH₃	CH ₃	C(O)C(CH ₃) ₃	CH ₃	
2.016	C≡CH	CH ₃	CH ₃	C(O)OCH ₂ CH ₃	CH ₃	
2.017	C≡CH	CH ₃	CH₂CH₃	Н	СН₃	
2.018	C≡CH	CH ₃	CH ₂ CH ₃	C(O)C(CH ₃) ₃	CH₃	
2.019	C≡CH	CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH ₃	
2.020	CH=CH ₂	CH ₃	CH=CH ₂	Н	СНз	
2.021	C≡CH	CH ₃	C≡CH	Н	СН₃	
2.022	OCH ₃	CH ₃	CH₂CH₃	Н	СН₃	
2.023	OCH ₃	CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃	СН₃	
2.024	OCH ₃	CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH ₃	
2.025	OCH ₃	CH ₃	Br	Н	CH_3	
2.026	OCH ₃	CH ₃	Br	$C(O)C(CH_3)_3$	CH ₃	
2.027	OCH ₃	CH ₃	Br	C(O)OCH ₂ CH ₃	СН3	
2.028	OCH ₃	CH ₃	C≡CH	Н	CH ₃	
2.029	OCH ₃	CH ₃	C≡CH	$C(O)C(CH_3)_3$	СН3	
2.030	OCH ₃	CH ₃	C≡CH	C(O)OCH ₂ CH ₃	СН3	
2.031	CH₃	C≡CH	CH ₃	н	CH ₃	
2.032	CH₂CH₃	C≡CH	CH ₃	Н	СН₃	

Table 3: Compounds of the formula Ig:

$$R_2 \xrightarrow{R_1} N \xrightarrow{N_{19}} (Ig)$$

Comp.	R ₁	R_2	R₃	G	R ₁₉	Phys. data
No.	CH ₃	CH₃	CH ₃	Н	СН₃	
3.001	CH₃	CH ₃	CH₃	C(O)C(CH ₃) ₃	СН₃	
3.002	CH₃	CH ₃	CH ₃		_	
3.003	CH ₂ CH ₃	CH₃	CH ₃	H	CH₃	
3.004		CH ₃			CH₃	
3.005		-	CH₂CH₃	H	CH₃	
3.006		CH₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH₃	
3.007	CH₂CH₃	CH₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH₃	
3.008	CH₂CH₃	CH₃	Br	Н	CH₃	
3.009	CH₂CH₃	CH₃	Br	C(O)C(CH ₃) ₃	CH₃	
3.010	CH₂CH₃	CH₃	Br	C(O)OCH₂CH₃	CH₃	
3.011	CH ₂ CH ₃	CH₂CH₃	CH₂CH₃	Н	CH ₃	
3.012	CH₂CH₃	CH₂CH ₃	CH₂CH₃	$C(O)C(CH_3)_3$	CH ₃	
3.013	CH₂CH₃	CH ₂ CH ₃	CH₂CH₃	C(O)OCH₂CH₃	CH₃	
3.014	C≡CH	CH₃	CH ₃	Н	CH_3	
3.015	C≡CH	CH ₃	CH₃	$C(O)C(CH_3)_3$	CH ₃	
3.016	C≡CH	CH ₃	CH₃	C(O)OCH ₂ CH ₃	CH ₃	
3.017	C≡CH	CH ₃	CH₂CH₃	Н	CH ₃	
3.018	C≡CH	CH ₃	CH₂CH₃	$C(O)C(CH_3)_3$	CH ₃	
3.019	C≡CH	CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH ₃	
3.020	CH=CH ₂	CH ₃	CH=CH ₂	н	СН3	
3.021	C≡CH	CH ₃	C≡CH	Н	CH ₃	
3.022	OCH ₃	CH ₃	CH ₂ CH ₃	Н	CH ₃	
3.023	OCH₃	CH ₃	CH ₂ CH ₃	$C(O)C(CH_3)_3$	CH ₃	
3.024	OCH ₃	CH₃	CH₂CH₃	C(O)OCH₂CH₃	СН₃	
3.025	OCH ₃	CH ₃	Br	Н	CH ₃	

Comp.	R ₁	R ₂	R ₃	G	R ₁₉	Phys. data
No.						
3.026	OCH ₃	CH ₃	Br	$C(O)C(CH_3)_3$	CH₃	
3.027	OCH ₃	CH ₃	Br	C(O)OCH ₂ CH ₃	CH₃	
3.028	OCH ₃	CH ₃	C≡CH	Н	CH ₃	
3.029	OCH ₃	CH ₃	C≡CH	$C(O)C(CH_3)_3$	CH ₃	
3.030	OCH ₃	CH ₃	C≡CH	C(O)OCH ₂ CH ₃	CH ₃	
3.031	CH ₃	C≡CH	CH₃	Н	CH ₃	
3.032	CH₂CH₃	C≡CH	CH ₃	Н	CH ₃	
3.033	CH₂CH₃	CH ₃	CH₂CH₃	Н	F	
3.034	CH ₂ CH ₃	CH ₃	CH ₂ CH ₃	Н	Br	
3.035	CH₃	CH ₃	CH ₃	Н	CI	
3.036	CH ₃	CH ₃	CH ₃	$C(O)C(CH_3)_3$	CI	
3.037	CH ₃	CH ₃	CH ₃	C(O)OCH ₂ CH ₃	CI	
3.038	CH ₂ CH ₃	CH₃	CH ₂ CH ₃	Н	CI	
3.039	CH₂CH₃	CH ₃	CH₂CH₃	$C(O)C(CH_3)_3$	CI	
3.040	CH₂CH₃	CH ₃	CH ₂ CH ₃	C(O)OCH ₂ CH ₃	CI	
3.041	CH ₂ CH ₃	CH ₂ CH ₃	CH ₂ CH ₃	Н	CI	
3.042	C≡CH	CH ₃	CH ₃	Н	CI	
3.043	C≡CH	CH ₃	C≡CH	Н	CI	
3.044	CH ₃	C≡CH	CH ₃	Н	CI	

Table 4: Compounds of the formula Ih:

$$R_{2}$$

$$R_{3}$$

$$G$$

$$R_{20}$$

$$R_{21}$$

$$R_{21}$$

$$R_{20}$$

$$R_{21}$$

$$R_{21}$$

$$R_{3}$$

$$R_{3}$$

$$R_{3}$$

$$R_{4}$$

$$R_{5}$$

Comp.	R_1	R_2	R_3	G	R ₂₀	R ₂₁	Phys.
No. 4.001	CH₃	CH₃	СН₃	Н	CH₃	CH ₃	data
4.002	CH ₃	CH ₃	CH₃	C(O)C(CH ₃) ₃	CH ₃	CH ₂	

Comp.	R ₁	R ₂	R ₃	G	R ₂₀	R ₂₁	Phys.
No. 4.003	СН₃	СН₃	СН₃	C(O)OCH ₂ CH ₃	CH₃	СН₃	data
4.003	CH₂CH₃	CH ₃	CH₂CH₃	Н	CH₃	CH₃	
4.005	CH₂CH₃	CH₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH₃	CH₃	
4.006	CH₂CH₃	CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH₃	CH ₃	
4.007	CH₂CH₃	CH₂CH₃	CH₂CH₃	Н	CH₃	CH₃	
4.008	CH₂CH₃	CH₂CH₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH ₃	CH₃	
4.009	CH₂CH₃	CH₂CH₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH ₃	CH₃	
4.010	C≡CH	CH ₃	CH ₃	Н	CH ₃	CH₃	
4.011	C≡CH	CH ₃	CH ₃	C(O)C(CH ₃) ₃	СН₃	CH₃	
4.012	C≡CH	CH ₃	CH₃	C(O)OCH ₂ CH ₃	CH₃	СН₃	
4.013	C≡CH	CH ₃	C≡CH	Н	CH ₃	СН₃	
4.014	CH ₃	C≡CH	CH ₃	н	CH ₃	CH₃	
4.015	CH ₃	CH ₃	CH ₃	н	CH ₂	CH₂	
4.016	CH₂CH₃	CH ₃	CH₂CH₃	н	CH₂	CH₂	
4.017	CH₂CH₃	CH₂CH₃	CH₂CH₃	Н	CH₂	CH₂	
4.018	CH₂CH₃	CH₃	CH₂CH₃	н	CH₂CI	H ₂ CH ₂	
4.019	CH ₃	CH ₃	CH ₃	н	CH₂CH₂	CH₂CH₂	
4.020	CH₂CH₃	CH ₃	CH₂CH₃	н	CH₂CH₂	CH₂CH₂	
4.021	CH₂CH₃	CH₂CH₃	CH₂CH₃	Н	CH₂CH₂	CH₂CH₂	

Comp. No.	R ₁	R_2	R ₃	G	R ₂₀	R ₂₁	Phys. data	
4.022	C≡CH	CH ₃	CH₃	Н	CH₂CH ₂			
4.023	C≡CH	CH₃	C≡CH	Н	CH₂CH ₂			
4.024	CH ₃	C≡CH	СН₃	Н	CH ₂ CH ₂ CH ₂ CH ₂			
4.025	CH₃	CH₃	CH₃	Н	CH₂CH₂C	H ₂ CH ₂ CH ₂		
4.026	CH₂CH₃	СН₃	CH₂CH₃	Н	CH₂CH₂C	H₂CH₂CH₂		
4.027	CH₂CH₃	CH₂CH₃	CH₂CH₃	Н	CH₂CH₂C	H ₂ CH ₂ CH ₂		
4.028	C≡CH	CH₃	CH₃	Н	CH₂CH₂C	H ₂ CH ₂ CH ₂		
4.029	C≅CH	CH ₃	C≡CH	н	CH₂CH₂C	H₂CH₂CH₂		
4.030	СН₃	C=CH	CH ₃	Н	CH₂CH₂C	H ₂ CH ₂ CH ₂		
4.031	CH₂CH₃	CH₃	CH₂CH₃	н	CH₂CH₂(OCH₂CH₂		

Table 5: Compounds of the formula lk:

Comp.	R_1	R ₂	R ₃	G	R ₁₈	R ₁₉	Phys.
No. 5.001	СН₃	CH₃	CH₃	. H	СН₃	CH₃	data
5.002	CH ₃	CH ₃	CH ₃	C(O)C(CH ₃) ₃	CH ₃	CH₃	
5.002	CH ₃	CH₃	CH₃		CH ₃	CH₃	
5.004	CH₂CH₃	CH₃	CH₂CH₃	H	CH ₃	CH ₃	
5.005	CH₂CH₃	СН₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH ₃	CH ₃	
5.006	CH₂CH₃	СН₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH₃	CH₃	
5.007	CH₂CH₃	CH₂CH₃	CH₂CH₃	Н	CH₃	CH₃	
5.008	CH₂CH₃	CH₂CH₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH₃	CH₃	
5.009	CH₂CH₃	CH₂CH₃	CH₂CH₃	C(O)OCH₂CH₃	CH₃	CH₃	
5.010	C≡CH	CH ₃	CH ₃	Н	CH ₃	CH₃	
5.011	C≡CH	СН₃	CH ₃	C(O)C(CH ₃) ₃	СН₃	СН₃	
5.012	C≡CH	CH ₃	CH ₃	C(O)OCH ₂ CH ₃	CH₃	CH₃	
5.013	C≡CH	CH ₃	C≡CH	Н	CH₃	CH₃	
5.014	C≝CH	СН₃	C≡CH	C(O)C(CH ₃) ₃	CH₃	СН₃	
5.015	C≡CH	CH ₃	C≡CH	C(O)OCH ₂ CH ₃	CH ₃	CH ₃	
5.016	CH₃	C≡CH	CH ₃	Н	CH ₃	CH₃	
5.017	CH ₃	CH ₃	CH ₃	Н	CH	I₂CH₂	
5.018	CH₂CH₃	CH ₃	CH₂CH₃	Н		I₂CH₂	
5.019	CH₂CH₃	CH₂CH₃	CH₂CH₃	Н	CH	l₂CH₂	
5.020	CH₂CH₃	CH ₃	CH₂CH₃	Н	CH₂C	CH₂CH₂	
5.021	CH ₃	CH ₃	CH ₃	Н	CH₂CH		
5.022	CH₂CH₃	CH ₃	CH₂CH₃	н			
5.023	CH₂CH₃	CH ₂ CH ₃	CH₂CH₃	Н			
5.024	C≡CH	CH ₃	CH ₃	Н			
5.025	C≡CH	CH ₃	C≡CH	Н	CH₂CH	₂ CH ₂ CH ₂	

Comp. No.	R ₁	R ₂	R ₃	G	R ₁₈	R ₁₉	Phys.
5.026	CH₃	C≅CH	CH ₃	Н	CH₂C	H ₂ CH ₂ CH ₂	data
5.027	CH ₃	CH ₃	CH ₃	Н	CH ₂ CH ₂	CH2CH2CH2	
5.028	CH₂CH₃	CH ₃	CH ₂ CH ₃	Н	CH₂CH₂	CH2CH2CH2	
5.029	CH₂CH₃	CH₂CH₃		Н	CH ₂ CH ₂	CH ₂ CH ₂ CH ₂	
5.030	C≡CH	CH ₃	CH ₃	Н	CH₂CH₂	CH ₂ CH ₂ CH ₂	
5.031	C≡CH	CH ₃	C≝CH	Н	CH ₂ CH ₂		
5.032	CH₃	C≡CH	CH ₃	Н	CH ₂ CH ₂	CH ₂ CH ₂ CH ₂	
5.033	CH ₂ CH ₃	CH ₃	CH₂CH₃	Н	CH₂C⊦	I₂OCH₂CH₂	

Table 6: Compounds of the formula Im:

$$R_{2}$$

$$R_{3}$$

$$R_{3}$$

$$R_{3}$$

$$R_{3}$$

$$R_{4}$$

$$R_{19}$$

$$R_{19}$$

$$R_{19}$$

$$R_{19}$$

Comp.	R ₁	R_2	R_3	G	R ₂₁	R ₁₉	Phys.
No.						13	data
6.001	CH ₃	CH ₃	CH ₃	Н	CH ₃	CH ₃	dulu
6.002	CH ₃	CH ₃	CH ₃	C(O)C(CH ₃) ₃	CH ₃	CH ₃	
6.003	CH₃	CH ₃	CH ₃	C(O)OCH ₂ CH ₃	CH ₃	CH ₃	
6.004	CH₂CH₃	CH ₃	CH ₂ CH ₃	Н	CH ₃	CH ₃	
6.005	CH₂CH₃	CH ₃	CH ₂ CH ₃	C(O)C(CH ₃) ₃	CH ₃	CH ₃	
6.006	CH₂CH₃	CH ₃	CH ₂ CH ₃	C(O)OCH ₂ CH ₃	CH₃	CH ₃	
6.007	CH₂CH₃	CH₂CH₃	CH₂CH₃	Н	CH₃	CH ₃	
6.008	CH₂CH₃	CH ₂ CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH ₃	CH ₃	
6.009	CH₂CH₃	CH₂CH₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH ₃	CH ₃	
6.010	C≡CH	CH ₃	CH ₃	Н	CH₃	CH ₃	
6.011	C≅CH	CH ₃	СНз	C(O)C(CH ₃) ₃	CH ₃	CH ₃	
6.012	C≡CH	CH ₃	CH ₃	C(O)OCH ₂ CH ₃	CH ₃	CH₃	
6.013	C≡CH	CH ₃	C≡CH	Н	CH ₃	СН₃	

Comp.	R_1	R ₂	R_3	G	R ₂₁	R ₁₉	Phys.
No.	0.011						data
6.014	C≡CH	CH₃	C≡CH	$C(O)C(CH_3)_3$	CH₃	CH₃	
6.015	C≡CH	CH ₃	C≡CH	C(O)OCH ₂ CH ₃	CH ₃	СН3	
6.016	CH ₃	C≡CH	CH ₃	Н	CH ₃	CH ₃	
6.017	CH ₂ CH ₃	CH ₃	CH₂CH₃	Н	CH₂C	H ₂ CH ₂	
6.018	CH ₂ CH ₃	CH ₃	CH ₂ CH ₃	Н	CH₂(OCH₂	
6.019	CH₂CH₃	CH₃	CH ₂ CH ₃	Н	CH₂CH₂	CH2CH2	

Table 7: Compounds of the formula in:

$$R_{2} \xrightarrow{R_{1} O R_{21}} O (In)$$

Comp.	R_1	R ₂	R ₃	G	R ₂₁	R ₁₅	Phys.
No.							data
7.001	CH ₃	CH₃	CH ₃	Н	CH₃	CH ₃	
7.002	CH₃	CH₃	CH₃	C(O)C(CH ₃) ₃	CH₃	CH₃	
7.003	CH₃	CH₃	CH ₃	C(O)OCH ₂ CH ₃	CH ₃	CH ₃	
7.004	CH₂CH₃	CH ₃	CH ₃	Н	CH ₃	CH ₃	
7.005	CH₂CH₃	CH ₃	CH ₃	$C(O)C(CH_3)_3$	CH ₃	CH ₃	
7.006	CH₂CH₃	CH ₃	CH ₃	C(O)OCH ₂ CH ₃	CH ₃	CH ₃	
7.007	CH₂CH₃	CH ₃	CH₂CH₃	H	CH ₃	CH₃	solid
7.008	CH₂CH₃	CH ₃	CH ₂ CH ₃	C(O)C(CH ₃) ₃	CH ₃	CH ₃	solid
7.009	CH₂CH₃	CH ₃	CH ₂ CH ₃	C(O)OCH ₂ CH ₃	CH ₃	CH ₃	
7.010	CH₂CH₃	CH ₃	Br	Н	CH ₃	CH₃	
7.011	CH₂CH₃	CH ₃	Br	$C(O)C(CH_3)_3$	CH ₃	CH₃	
7.012	CH₂CH₃	CH ₃	Br	C(O)OCH ₂ CH ₃	CH ₃	CH ₃	
7.013	CH₂CH₃	CH ₂ CH ₃	CH₂CH₃	Н	CH ₃	CH ₃	
7.014	CH₂CH₃	CH ₂ CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH ₃	CH ₃	
7.015	CH₂CH₃	CH ₂ CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH₃	CH ₃	
7.016	C≡CH	CH ₃	CH₃	Н	CH₃	CH ₃	
7.017	C≡CH	CH ₃	CH ₃	$C(O)C(CH_3)_3$	CH ₃	CH ₃	
7.018	C≣CH	CH ₃	CH₃	C(O)OCH ₂ CH ₃	CH₃	CH ₃	
7.019	C≡CH	CH ₃	CH₂CH₃	Н	CH ₃	CH ₃	
7.020	C≡CH	CH ₃	CH₂CH₃	$C(O)C(CH_3)_3$	CH ₃	CH ₃	
7.021	C≡CH	CH ₃	CH ₂ CH ₃	C(O)OCH ₂ CH ₃	CH ₃	CH ₃	
7.022	CH=CH ₂	CH ₃	CH=CH ₂	Н	CH ₃	CH ₃	
7.023	C≡CH	CH ₃	C≡CH	Н	CH ₃	CH ₃	
7.024	C≡CH	CH ₃	C≡CH	$C(O)C(CH_3)_3$	CH ₃	CH ₃	

Comp.	R_1	R_2	R_3	G	R ₂₁	R ₁₅	Phys.
No.	C≡CH	CH₃	C-CH	C(O)OCH CH	СH	CH	data
7.025	O≘CH ₃		C≡CH	C(O)OCH₂CH₃	CH₃	CH₃	
7.026	OCH₃ OCH₃	CH₃	CH₂CH₃	H	CH₃	CH₃	
7.027	OCH₃ OCH₃	CH₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH₃	CH₃	
7.028		CH₃	CH₂CH₃	C(O)OCH₂CH₃	CH₃	CH₃	
7.029	OCH₃	CH₃	Br	H	CH₃	CH₃	
7.030	OCH₃	CH₃	Br	C(O)C(CH ₃) ₃	CH₃	CH₃	
7.031	OCH₃	CH₃	Br CCU	C(O)OCH₂CH₃	CH₃	CH₃	
7.032	OCH₃	CH₃	C≡CH	Н	CH₃	CH₃	
7.033	OCH ₃	CH ₃	C≡CH	C(O)C(CH ₃) ₃	CH ₃	CH₃	
7.034	OCH ₃	CH₃	C≡CH	C(O)OCH ₂ CH ₃	CH₃	CH₃	
7.035	CH₃	C≡CH	CH₃	Н	CH ₃	CH₃	
7.036	CH₂CH₃	C≡CH	CH ₃	Н	СН₃	CH₃	
7.037	CH ₃	CH ₃	CH ₃	Н		CH₂	
7.038	CH₃	CH₃	CH ₃	C(O)C(CH ₃) ₃		CH ₂	
7.039	CH ₃	CH ₃	CH ₃	C(O)OCH ₂ CH ₃		CH ₂	
7.040	CH₂CH₃	CH ₃	CH₂CH₃	н		CH₂	
7.041	CH₂CH₃	CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃		CH ₂	
7.042	CH₂CH₃	CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃		CH ₂	
7.043	CH₂CH₃	CH ₃	Br	н		CH ₂	
7.044	CH₂CH₃	CH ₃	Br	C(O)C(CH ₃) ₃		CH ₂	
7.045	CH₂CH₃	CH ₃	Br	C(O)OCH ₂ CH ₃		CH ₂	
7.046	С≡СН	CH ₃	CH ₃	Н		CH ₂	

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Comp.	R ₁	R_2	R ₃	G	R ₂₁	R ₁₅	Phys.
No. 7.047	C≅CH	CH₃	CH ₃	C(O)C(CH ₃) ₃		CH₂	data
7.048	C≡CH	CH₃	CH ₃	C(O)OCH₂CH₃		CH ₂	
7.049	C≡CH	CH₃	CH₂CH₃	н		CH ₂	
7.050	C≡CH	CH₃	CH₂CH₃	C(O)C(CH ₃) ₃		CH ₂	
7.051	C≡CH	CH ₃	CH₂CH₃	C(O)OCH₂CH₃		CH₂	
7.052	C≡CH	СН₃	C≡CH	Н		CH₂	
7.053	C≡CH	CH ₃	C≡CH	C(O)C(CH ₃) ₃		CH₂	
7.054	C≡CH	CH₃	C≡CH	C(O)OCH₂CH₃		CH₂	
7.055	OCH₃	СН₃	CH₂CH₃	Н		CH₂	
7.056	OCH ₃	CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃		CH₂	
7.057	OCH ₃	CH₃	CH₂CH₃	C(O)OCH ₂ CH ₃		CH ₂	
7.058	OCH ₃	СН₃	Br	Н		CH₂	
7.059	OCH ₃	СН₃	Br	C(O)C(CH ₃) ₃		CH₂	
7.060	OCH ₃	CH ₃	Br	C(O)OCH ₂ CH ₃		CH ₂	
7.061	OCH ₃	CH ₃	C≡CH	Н		CH ₂	
7.062	OCH ₃	CH ₃	C≡CH	C(O)C(CH ₃) ₃		CH ₂	
7.063	OCH ₃	CH ₃	C≡CH	C(O)OCH ₂ CH ₃		CH ₂	

Comp. No.	R ₁	R ₂	R ₃	G	R ₂₁	R ₁₅	Phys. data
7.064	CH ₃	CH ₃	CH ₃	Н		CH ₂ CH ₂	
7.065	CH ₃	CH ₃	CH ₃	C(O)C(CH ₃) ₃		CH ₂ CH ₂	
7.066	CH₃	CH ₃	CH₃	C(O)OCH ₂ CH ₃		CH₂ CH₂	
7.067	CH₂CH₃	CH ₃	CH₂CH₃	Н		CH ₂ CH ₂	
7.068	CH₂CH₃	CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃		CH ₂ CH ₂	
7.069	CH₂CH₃	CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃		CH ₂ CH ₂	
7.070	CH₂CH₃	CH₃	Br	н		CH ₂ CH ₂	
7.071	CH₂CH₃	CH ₃	Br	C(O)C(CH ₃) ₃		CH ₂ CH ₂	
7.072	CH₂CH₃	CH ₃	Br	C(O)OCH ₂ CH ₃		CH ₂ CH ₂	
7.073	C≡CH	CH ₃	CH₃	Н		CH ₂ CH ₂	
7.074	C≡CH	CH ₃	CH₃	C(O)C(CH ₃) ₃		CH ₂ CH ₂	
7.075	C≅CH	CH₃	CH₃	C(O)OCH ₂ CH ₃		CH ₂ CH ₂	
7.076	C≡CH	CH ₃	CH₂CH₃	Н		CH ₂ CH ₂	
7.077	C≡CH	CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃		CH ₂ CH ₂	
7.078	C≡CH	CH₃	CH₂CH₃	C(O)OCH₂CH₃		CH₂ CH₂	
7.079	C≡CH	CH ₃	C≡CH	Н		CH ₂ CH ₂	

Comp. No.	R ₁	R ₂	R ₃	G	R ₂₁	R ₁₅	Phys.
7.080	C≡CH	CH₃	C≡CH	C(O)C(CH ₃) ₃		CH ₂ CH ₂	data
7.081	C≡CH	CH ₃	C≡CH	C(O)OCH ₂ CH ₃		CH ₂ CH ₂	
7.082	OCH ₃	CH ₃	CH₂CH₃	Н		CH ₂ CH ₂	
7.083	OCH ₃	CH₃	CH₂CH₃	C(O)C(CH ₃) ₃		CH ₂ CH ₂	
7.084	OCH ₃	CH ₃	CH₂CH₃	C(O)OCH₂CH₃		CH ₂ CH ₂	
7.085	OCH ₃	CH ₃	Br	Н		CH ₂ CH ₂	
7.086	OCH ₃	CH₃	Br	C(O)C(CH ₃) ₃		CH ₂ CH ₂	
7.087	OCH₃	CH ₃	Br	C(O)OCH₂CH₃		CH₂ CH₂	
7.088	OCH₃	CH ₃	C≡CH	н		CH ₂ CH ₂	
7.089	OCH ₃	CH ₃	C≡CH	C(O)C(CH ₃) ₃		CH ₂ CH ₂	
7.090	OCH ₃	CH ₃	C≡CH	C(O)OCH₂CH₃		CH ₂ CH ₂	

Table 8: Compounds of the formula lo:

Comp.	R_1	R_2	R_3	G	R ₁₉	R ₁₇	Phys.
No.						.,	data

Comp.	R ₁	R_2	R ₃	G	R ₁₉	R ₁₇	Phys.
No.	СН₃	СН₃	CH₃	Н	CH	CH	data
8.001	CH₃	CH₃ CH₃	CH₃	C(O)C(CH ₃) ₃	CH₃ CH₃	CH₃	
8.002	CH₃	CH ₃	CH₃		CH₃ CH₃	CH₃ CH₃	
8.003 8.004	CH₂CH₃	CH₃	CH ₃	H	CH ₃	CH ₃	
8.004	CH₂CH₃	CH₃	CH₃	C(O)C(CH ₃) ₃	CH ₃	CH₃	
8.006	CH₂CH₃	CH₃	CH₃	C(O)OCH ₂ CH ₃	CH ₃	CH₃	
8.007	CH₂CH₃	CH₃	CH₂CH₃	H	CH ₃	CH ₃	
8.008	CH₂CH₃	CH₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH ₃	CH₃	
8.009	CH₂CH₃	СН₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH₃	CH₃	
8.010	CH₂CH₃	CH₃	Br	Н	CH₃	CH₃	
8.011	CH₂CH₃	CH ₃	Br	C(O)C(CH ₃) ₃	CH₃	CH₃	
8.012	CH ₂ CH ₃	CH ₃	Br	C(O)OCH₂CH₃	СН₃	CH ₃	
8.013	CH ₂ CH ₃	CH ₂ CH ₃	CH₂CH₃	Н	СН₃	СН₃	
8.014	CH ₂ CH ₃	CH₂CH₃	CH₂CH₃	$C(O)C(CH_3)_3$	СНз	СН₃	
8.015	CH₂CH₃	CH ₂ CH ₃	CH₂CH₃	C(O)OCH₂CH₃	СНз	CH₃	
8.016	C≡CH	CH ₃	CH ₃	Н	СН₃	СН3	
8.017	C≣CH	CH ₃	CH ₃	$C(O)C(CH_3)_3$	СН₃	СН₃	
8.018	C≡CH	СН₃	CH ₃	C(O)OCH ₂ CH ₃	СНз	CH ₃	
8.019	C≡CH	CH ₃	CH₂CH₃	Н	СН₃	CH₃	
8.020	C≡CH	CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH ₃	CH ₃	
8.021	C≡CH	CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH ₃	СН₃	
8.022	CH=CH ₂	CH ₃	CH=CH ₂	Н	CH₃	СН₃	
8.023	C≡CH	CH₃	C≡CH	Н	CH ₃	CH₃	
8.024	C≡CH	CH ₃	C≡CH	C(O)C(CH ₃) ₃	CH ₃	CH ₃	,
8.025	C≡CH	CH ₃	C≡CH	C(O)OCH ₂ CH ₃	CH₃	CH ₃	
8.026	OCH ₃	CH ₃	CH₂CH₃	Н	CH₃	СН₃	
8.027	OCH ₃	CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃	CH₃	СН₃	
8.028	OCH ₃	CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃	CH ₃	CH ₃	
8.029	OCH ₃	CH ₃	Br	Н	CH₃	CH ₃	
8.030	OCH ₃	CH ₃	Br	C(O)C(CH ₃) ₃	CH ₃	СН₃	

Comp.	R ₁	R_2	R_3	G	R ₁₉	R ₁₇	Phys.
No. 8.031	OCH₃	СН₃	Br	C(O)OCH₂CH₃	CH ₃	CH₃	data
8.032	OCH ₃	CH₃	C≣CH	H	CH ₃	CH ₃	
8.033	OCH ₃	CH ₃	C≡CH	C(O)C(CH ₃) ₃	CH₃	CH₃	
8.034	OCH ₃	СНз	C≡CH	C(O)OCH ₂ CH ₃	CH₃	CH ₃	
8.035	CH ₃	C≡CH	CH₃	н	СН₃	CH₃	
8.036	CH₂CH₃	C≝CH	CH₃	Н	СН₃	CH₃	
8.037	CH₂CH₃	CH₃	CH₂CH₃	Н	F	F	
8.038	CH ₃	СН₃	CH ₃	Н	CI	CI	
8.039	CH ₂ CH ₃	CH ₃	CH ₂ CH ₃	Н	CI	CI	
8.040	CH ₃	CH ₃	CH₃	Н		CH ₂ CH ₂	m.p. 295°C
8.041	CH ₃	CH ₃	CH ₃	$C(O)C(CH_3)_3$		CH ₂ CH ₂	m.p.
							198-
							199°C
8.042	CH ₃	CH ₃	CH ₃	C(O)OCH ₂ CH ₃		CH ₂ CH ₂	
8.043	CH ₂ CH ₃	CH ₃	CH₂CH ₃	Н		CH ₂ CH ₂	m.p.
	011 011						287°C
8.044	CH₂CH₃	CH₃	CH₂CH₃	C(O)C(CH ₃) ₃		CH ₂ CH ₂	m.p.
							141-
	CH₂CH₃	CH₃	CH CH	C(O)OOU OU			143°C
8.045	CH ₂ CH ₃	CH₃ CH₃	CH₂CH₃ Br	C(O)OCH₂CH₃		CH₂CH₂	
8.046		CH₃ CH₃	Br	H C(O)C(CH ₃) ₃		CH₂CH₂	
8.047		CH₃ CH₃	Br			CH₂CH₂	
8.048	C≣CH	CH₃	CH₃	C(O)OCH₂CH₃ H		CH ₂ CH ₂	
8.049	C≡CH	CH₃	CH₃	C(O)C(CH ₃) ₃		CH₂CH₂	
8.050	C≡CH		-			CH₂CH₂	
8.051		CH₃	CH₃	C(O)OCH ₂ CH ₃		CH₂CH₂	
8.052	C≣CH	CH₃	CH₂CH₃	Н		CH ₂ CH ₂	
8.053	C≡CH	CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃		CH ₂ CH ₂	
8.054	C≡CH	CH₃	CH₂CH₃	C(O)OCH ₂ CH ₃		CH ₂ CH ₂	

Comp.	R ₁	R_2	R ₃	G	R ₁₉	R ₁₇	Phys.
No. 8.055	C≡CH	СН₃	C≡CH	Н		CH₂CH₂	data
8.056	C≡CH	CH₃	C≡CH	C(O)C(CH ₃) ₃		CH ₂ CH ₂	
8.057	C≡CH	CH ₃	C≡CH	C(O)OCH ₂ CH ₃		 CH₂CH₂	
8.058	OCH₃	CH₃	CH₂CH₃	Н		 CH₂CH₂	
8.059	OCH ₃	CH₃	CH₂CH₃	C(O)C(CH ₃) ₃		CH₂CH₂	
8.060	OCH ₃	CH₃	CH₂CH₃	C(O)OCH ₂ CH ₃		CH₂CH₂	
8.061	OCH ₃	CH ₃	Br	Н		CH₂CH₂	
8.062	OCH ₃	CH ₃	Br	C(O)C(CH ₃) ₃		CH₂CH₂	
8.063	OCH ₃	CH ₃	Br	C(O)OCH₂CH₃		CH₂CH₂	
8.064	OCH ₃	CH ₃	C≡CH	Н		CH ₂ CH ₂	
8.065	OCH ₃	CH ₃	C≡CH	C(O)C(CH ₃) ₃		CH₂CH₂	
8.066	OCH ₃	CH₃	C≡CH	C(O)OCH ₂ CH ₃		CH₂CH₂	
8.067	CH ₃	CH ₃	CH₃	Н		CH₂CH₂CH₂	
8.068	CH ₃	CH ₃	CH ₃	$C(O)C(CH_3)_3$		CH₂CH₂CH₂	
8.069	CH₃	CH ₃	CH ₃	C(O)OCH₂CH₃		CH₂CH₂CH₂	
8.070	CH₂CH₃	CH ₃	CH₂CH₃	Н		CH₂CH₂CH₂	
8.071	CH₂CH₃	CH ₃	CH ₂ CH ₃	$C(O)C(CH_3)_3$		CH₂CH₂CH₂	
8.072	CH₂CH₃	CH ₃	CH ₂ CH ₃	C(O)OCH ₂ CH ₃		CH₂CH₂CH₂	
8.073	CH₂CH₃	CH_3	Br	Н		CH₂CH₂CH₂	
8.074	CH₂CH₃	CH ₃	Br	$C(O)C(CH_3)_3$		CH₂CH₂CH₂	
8.075	CH₂CH₃	CH ₃	Br	C(O)OCH ₂ CH ₃	,	CH₂CH₂CH₂	
8.076	C≡CH	CH ₃	CH₃	Н		CH₂CH₂CH₂	
8.077	C≡CH	CH ₃	CH ₃	C(O)C(CH ₃) ₃		CH₂CH₂CH₂	
8.078	C≡CH	CH ₃	CH₃	C(O)OCH ₂ CH ₃	ı	CH₂CH₂CH₂	
8.079	C≡CH	CH ₃	CH₂CH₃	Н	•	CH₂CH₂CH₂	
8.080	C≡CH	CH ₃	CH₂CH₃	C(O)C(CH ₃) ₃	(CH₂CH₂CH₂	
8.081	C≡CH	CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃	(CH₂CH₂CH₂	
8.082	C≡CH	СН₃	C≡CH	Н	(CH ₂ CH ₂ CH ₂	
8.083	C≡CH	СН₃	C≡CH	C(O)C(CH ₃) ₃	•	CH ₂ CH ₂ CH ₂	
8.084	C≡CH	СН₃	C≡CH	C(O)OCH₂CH₃	(
						- -	

Comp.	R ₁	R ₂	R ₃	G	R ₁₉	R ₁₇	Phys.
No. 8.085	OCH₃	СН₃	CH₂CH₃	Н	C		data
8.086	OCH ₃	CH₃	CH₂CH₃	C(O)C(CH ₃) ₃		CH ₂ CH ₂ CH ₂	
8.087	OCH ₃	CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃			
8.088	OCH ₃	CH₃	Br	Н	C	CH₂CH₂CH₂	
8.089	OCH ₃	CH ₃	Br	$C(O)C(CH_3)_3$	C	CH₂CH₂CH₂	
8.090	OCH ₃	CH_3	Br	C(O)OCH ₂ CH ₃	C	CH₂CH₂CH₂	
8.091	OCH ₃	CH ₃	C≡CH	Н	C	CH₂CH₂CH₂	
8.092	OCH ₃	CH ₃	C≡CH	$C(O)C(CH_3)_3$	C	CH₂CH₂CH₂	
8.093	OCH ₃	CH ₃	C≡CH	C(O)OCH ₂ CH ₃	C	CH₂CH₂CH₂	
8.094	CH ₃	CH ₃	CH₃	н		CH ₂ OCH ₂	
8.095	CH₃	CH ₃	CH₃	$C(O)C(CH_3)_3$		CH ₂ OCH ₂	
8.096	CH₃	CH ₃	CH₃	C(O)OCH ₂ CH ₃		CH ₂ OCH ₂	
8.097	CH₂CH₃	CH ₃	CH₂CH₃	Н		CH ₂ OCH ₂	
8.098	CH₂CH₃	CH ₃	CH₂CH₃	$C(O)C(CH_3)_3$		CH ₂ OCH ₂	
8.099	CH₂CH₃	CH₃	CH₂CH₃	C(O)OCH ₂ CH ₃		CH₂OCH₂	
8.100	CH₂CH₃	CH ₃	Br	Н		CH ₂ OCH ₂	
8.101	CH₂CH₃	CH ₃	Br	$C(O)C(CH_3)_3$		CH₂OCH₂	
8.102	CH₂CH₃	CH ₃	Br	C(O)OCH ₂ CH ₃		CH₂OCH₂	
8.103	C≡CH	CH ₃	CH ₃	Н		CH₂OCH₂	
8.104	C≡CH	CH ₃	CH ₃	$C(O)C(CH_3)_3$		CH ₂ OCH ₂	
8.105	C≡CH	CH ₃	CH ₃	C(O)OCH ₂ CH ₃		CH ₂ OCH ₂	
8.106	C≡CH	CH ₃	CH₂CH₃	Н		CH ₂ OCH ₂	
8.107	C≡CH	CH ₃	CH₂CH₃	$C(O)C(CH_3)_3$		CH ₂ OCH ₂	
8.108	C≡CH	CH ₃	CH₂CH₃	C(O)OCH₂CH₃		CH ₂ OCH ₂	
8.109	C≅CH	CH ₃	C≡CH	Н		CH ₂ OCH ₂	
8.110	C≡CH	CH ₃	C≡CH	$C(O)C(CH_3)_3$		CH ₂ OCH ₂	
8.111	C≡CH	CH ₃	C≡CH	C(O)OCH ₂ CH ₃		CH ₂ OCH ₂	
8.112	OCH ₃	CH ₃	CH ₂ CH ₃	Н		CH ₂ OCH ₂	
8.113	OCH ₃	CH₃	CH ₂ CH ₃	$C(O)C(CH_3)_3$		CH ₂ OCH ₂	
8.114	OCH ₃	CH ₃	CH₂CH₃	C(O)OCH ₂ CH ₃		CH ₂ OCH ₂	

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Comp.	R ₁	R_2	R ₃	G	R ₁₉	R ₁₇	Phys.
No.	* *1	1.2	• • • •	u	1 119	117	data
8.115	OCH ₃	CH ₃	Br	Н	CI		data
8.116	OCH ₃	CH₃	Br	C(O)C(CH ₃) ₃	CI		
8.117	OCH ₃	CH₃	Br	C(O)OCH ₂ CH ₃	CI		
8.118	OCH ₃	CH₃	C≡CH	Н	CI		
8.119	OCH ₃	CH ₃	C≝CH	C(O)C(CH ₃) ₃	CI	H₂OCH₂	
8.120	OCH ₃	CH₃	C≡CH	C(O)OCH ₂ CH ₃	CI	H ₂ OCH ₂	
<u>Formulation</u>	on examples	for herbic	idally active o	compounds of the f	ormula l		
(% = per c	ent by weigl	<u>nt)</u>					
F1. Emuls	ion concentr	ates	a)	b)	c)		d)
Active con	npound acco	ording	5%	10%	25%		50%
to Tables	1-8						
Ca dodecy	/lbenzenesu	lfonate	6%	8%	6%		8%
	polyglycol et	her	4%	-	4%		4%
(36 mol of	EO)						
	ol polyglycol	ether	-	4%	-		2%
(7- 8 mol c	•						
Cyclohexa			-	-	10%		20%
Arom. hyd			85%	78%	55%		16%
mixture C ₉	· -						
with water		red conce	ntration can b	e prepared from su	ich conce	ntrates by	dilution
F2. Solution			a)	b)	c)		d)
	npound acco	rding	5%	10%	50%		90%
to Tables							
-	-3-(3-metho	xy-					
propoxy)pi	•		-	20%	20%		-
	ne giycol MV		20%	10%	-		-
_	2-pyrrolidone	1	-	-	30%		10%
Arom. hyd			75%	60%	-		-
mixture C ₉	-C ₁₂						

The solutions are suitable for use in the form of tiny droplets.

F3. Wettable powders	a)	b)	c)	d)
Active compound according	5%	25%	50%	80%
to Tables 1-8				
Sodium lignosulfonate	4%	-	3%	-
Sodium laurylsulfate	2%	3%	-	4%
Sodium diisobutylnaphthalene-	-	6%	5%	6%
sulfonate				
Octylphenol polyglycol ethe	-	1%	2%	-
(7-8 mol of EO)				
Finely divided silica	1%	3%	5%	10%
Kaolin	88%	62%	35%	-

The active compound is thoroughly mixed with the additives and ground well in a suitable mill. This gives spray powders which can be diluted with water to give suspensions of any desired concentration.

F4. Coated granules	a)	b)	c)
Active compound according	0.1%	5%	15%
to Tables 1-8			
Finely divided silica	0.9%	2%	2%
Inorg. carrier material	99.0%	93%	83%
(Æ 0.1 - 1 mm),			

for example CaCO₃ or SiO₂

The active compound is dissolved in methylene chloride, the solution is sprayed onto the carrier and the solvent is subsequently evaporated off under reduced pressure.

F5. Coated granules	a)	b)	c)
Active compound according	0.1%	5%	15%
to Tables 1-8			
Polyethylene glycol MW 200	1.0%	2%	3%
Finely divided silica	0.9%	1%	2%
Inorg. carrier material	98.0%	92%	80%
(Æ 0.1 - 1 mm),			

for example $CaCO_3$ or SiO_2

In a mixer, the finely ground active compound is applied evenly to the carrier material moistened with polyethylene glycol. In this manner, dust-free coated granules are obtained.

F6. Extruder granules	a)	b)	c)	d)
Active compound according	0.1%	3%	5%	15%
to Tables 1-8				
Sodium lignosulfonate	1.5%	2%	3%	4%
Carboxymethylcellulose	1.4%	2%	2%	2%
Kaolin	97.0%	93%	90%	79%

The active compound is mixed with the additives, ground and moistened with water. This mixture is extruded and subsequently dried in a stream of air.

F7. Dusts	a)	b)	c)
Active compound according	0.1%	1%	5%
to Tables 1-8			
Talc mixture	39.9%	49%	35%
Kaolin	60.0%	50%	60%

Ready-to-use dusts are obtained by mixing the active compound with the carriers and grinding the mixture in a suitable mill.

F8. Suspension concentrates	a)	b)	c)	d)
Active compound according	3%	10%	25%	50%
to Tables 1-8				32,5
Ethylene glycol	5%	5%	5%	5%
Nonylphenol polyglycol ether	-	1%	2%	0,0
-				
(15 mol of EO)				
Sodium lignosulfonate	3%	3%	4%	5%
Carboxymethylcellulose	1%	1%	1%	1%
37% aqueous formaldehyde	0.2%	0.2%	0.2%	0.2%
solution				
Silicone oil emulsion	0.8%	0.8%	0.8%	0.8%
Water	87%	79%	62%	38%

The finely ground active compound is intimately mixed with the additives. This gives a suspension concentrate, from which suspensions of any desired concentration can be prepared by dilution with water.

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Biological Examples

Experimental comparison with the prior art:

The following compounds were examined for their herbicidal activity:

Compound No. 1.01

according to the present invention, and compound A

from the prior art (EP-A-0 508 126, compound no. 46 of Table 1).

Example B1: Herbicidal action before emergence of the plants (pre-emergence action) Monocotyledonous and dicotyledonous weeds are sown in standard soil in plastic pots. Immediately after sowing, the test substances are applied (500 l of water/ha) as an aqueous suspension (prepared using a 25% wettable powder (Example F3, b)) or as an emulsion (prepared using a 25% emulsion concentrate (Example F1, c)). The application rate is 500 g of active substance/ha. The test plants are subsequently grown under optimum conditions in a greenhouse. 3 weeks after the application, evaluation is carried out using a nine-level scale of ratings (1 = complete damage, 9 = no effect). Ratings of 1 to 4 (in particular 1 to 3) mean a good to very good herbicidal action.

Test plants: Alopecurus (Alo), Avena (Ave), Lolium (Lol), Setaria (Set), Panicum (Pan), Sorghum (Sor), Digitaria (Dig), Echinocloa (Ech) and Brachiaria (Bra).

Table B1: Pre-emergence action:

Pre-emergence action at 500 g of ai/ha

Comp. No.	Alo	Ave	Lol	Set	Pan	Sor	Dig	Ech	Bra
Compound A	5	7	4	3	7	6	7	5	3
1.001	3	4	1	1	1	1	2	1	2

Example B2: Herbicidal action after emergence of the plants (post-emergence action): Monocotyledonous and dicotyledonous weeds are grown in standard soil in plastic pots under greenhouse conditions. The test substances are applied at the 3- to 6-leaf stage of the test plants. The test substances are applied (500 l of water/ha) as an aqueous suspension (prepared using a 25% wettable powder (Example F3, b)) or as an emulsion (prepared using a 25% emulsion concentrate (Example F1, c)) at an application rate of 500 g of active substance/ha. 3 weeks after the application, evaluation is carried out using a nine-level scale of ratings (1 = complete damage, 9 = no effect). Ratings of 1 to 4 (in particular 1 to 3) mean a good to very good herbicidal action.

Test plants: Alopecurus (Alo), Avena (Ave), Lolium (Lol), Setaria (Set), Panicum (Pan), Sorghum (Sor), Digitaria (Dig), Echinocloa (Ech) and Brachiaria (Bra).

Table B2: Post-emergence action:

Post-emergence action at 500 g of ai/ha

Comp. No.	Alo	Ave	Lol	Set	Pan	Sor	Dig	Ech	Bra
Compound A	5	2	5	4	2	3	5	1	2
1.001	2	1	1	1	1	1	1	1	1

Comparing the herbicidal action of the compound A of the prior art with the compound no. 1.01 of the present invention, it can be seen that the compound no. 1.01 surprisingly exhibits considerably better herbicidal action against all of the weeds tested, although this compound differs from the compound A only in that an alkylene group in the ring has been replaced by oxygen.

Example B3: Herbicidal action of compounds of the present invention before emergence of the plants (pre-emergence action):

Monocotyledonous and dicotyledonous weeds are grown in standard soil in plastic pots. Directly after sowing, the test substances are applied (500 l of water/ha) as an aqueous suspension (prepared using a 25% wettable powder (Example F3, b)) or as an emulsion (prepared using a 25% emulsion concentrate (Example F1, c)). The application rate is 500 g of active substance/ha. The test plants are subsequently grown under optimum conditions in a greenhouse 3 weeks after the application, evaluation is carried out using a nine-level

scale of ratings (1 = complete damage, 9 = no effect). Ratings of 1 to 4 (in particular 1 to 3) mean a good to very good herbicidal action.

Test plants: Avena (Ave), Lolium (Lol), Setaria (Set).

Table B3: Pre-emergence action:

		Test plant:	
Comp. No.	Ave	Lol	Set
1.001	4	1	1
1.008	1	1	1
1.004	1	1	2

The same results are obtained when the compounds of the formula I are formulated according to Examples F2 and F4 to F8.

Example B4: Herbicidal action of compounds of the present invention after emergence of the plants (post-emergence action):

Monocotyledonous and dicotyledonous weeds are grown in standard soil in plastic pots under greenhouse conditions. The test substances are applied at the 3- to 6-leaf stage of the test plants. The test substances are applied (500 l of water/ha) as an aqueous suspension (prepared using a 25% wettable powder (Example F3, b)) or as an emulsion (prepared using a 25% emulsion concentrate (Example F1, c)) at an application rate of 250 g of active substance/ha. 3 weeks after the application, evaluation is carried out using a nine-level scale of ratings (1 = complete damage, 9 = no effect). Ratings of 1 to 4 (in particular 1 to 3) mean a good to very good herbicidal action.

Test plants: Avena (Ave), Lolium (Lol), Setaria (Set).

Table B4: Post-emergence action;

	Test plant:				
Comp. No.	Ave	Lol	Set		

1.001	1	1	2
1.088	4	4	3
1.078	1	1	4
1.007	1	1	1
1.005	1	2	2
1.085	1	2	2
1.016	3	2	2

The same results are obtained when the compounds of the formula I are formulated according to Examples F2 and F4 to F8.

WHAT IS CLAIMED IS:

1. A compound of the formula I

$$R_{4}$$
 N_{1}
 R_{4}
 R_{4}
 R_{3}
 R_{4}
 R_{3}
 R_{4}
 R_{4}
 R_{5}
 R_{4}
 R_{5}
 R_{5}
 R_{5}
 R_{5}
 R_{2}
 R_{3}
 R_{4}
 R_{5}
 R_{5}

in which

 R_1 , R_2 and R_3 independently of one another are halogen, nitro, cyano, C_1 - C_4 alkyl, C_2 - C_4 alkenyl, C_2 - C_4 alkynyl, C_1 - C_4 haloalkyl, C_2 - C_6 haloalkenyl, C_3 - C_6 cycloalkyl, halogen-substituted C_3 - C_6 cycloalkyl, C_1 - C_6 alkoxyalkyl, C_1 - C_6 alkylthioalkyl, hydroxyl, mercapto, C_1 - C_6 alkoxy, C_3 - C_6 alkenyloxy, C_3 - C_6 alkynyloxy, C_1 - C_4 alkylcarbonyl, C_1 - C_4 alkoxycarbonyl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, amino, C_1 - C_4 alkylamino or di(C_1 - C_4 -alkyl)amino;

R₄ and R₅ together are a group

 $-C-R_6(R_7)-O-C-R_8(R_9)-C-R_{10}(R_{11})-C-R_{12}(R_{13})-$ (Z₁),

 $-C-R_{14}(R_{15})-C-R_{16}(R_{17})-O-C-R_{18}(R_{19})-C-R_{20}(R_{21})-$ (Z₂), or

 $-C-R_{22}(R_{23})-C-R_{24}(R_{25})-C-R_{26}(R_{27})-O-C-R_{28}(R_{29})-$ (Z₃);

in which R_6 , R_7 , R_8 , R_9 , R_{10} , R_{11} , R_{12} , R_{13} , R_{14} , R_{15} , R_{16} , R_{17} , R_{18} , R_{19} , R_{20} , R_{21} , R_{22} , R_{23} , R_{24} , R_{25} , R_{26} , R_{27} , R_{28} , and R_{29} independently of one another are hydrogen, halogen, C_1 - C_4 alkyl or C_1 - C_4 haloalkyl where an alkylene ring, which together with the carbon atoms of the groups Z_1 , Z_2 or Z_3 contains 2 to 6 carbon atoms and may be interrupted by oxygen, may either be fused or spiro-linked to the carbon atoms of the groups Z_1 , Z_2 or Z_3 , or where this alkylene ring bridges at least one ring atom of the groups Z_1 , Z_2 or Z_3 ; C_1 is hydrogen, C_1 - C_1 - C_2 - C_3 -

 X_1 , X_2 , X_3 , X_4 and X_5 independently of one another are oxygen or sulfur; and R_{30} , R_{31} , R_{32} , R_{33} , R_{34} , R_{35} and R_{36} independently of one another are hydrogen, C_1 - C_5 alkyl, C_1 - C_5 haloalkyl, C_2 - C_5 alkenyl, C_1 - C_5 alkoxyalkyl, C_3 - C_6 cycloalkyl or phenyl, and R_{34} is additionally C_2 - C_2 0alkenyl, C_2 - C_2 0alkenyl substituted by halogen, alkylcarbonyl, alkylcarbonyloxy, alkoxy, thioalkyl, alkylthiocarbonyl, alkylcarbonylthio, alkylsulfonyl, alkylsulfoxyl, alkylaminosulfonyl, dialkylaminosulfonyl, alkylsulfonyloxy,

alkylsulfonylamino, alkylamino, dialkylamino, alkylcarbonylamino, dialkylcarbonylamino, alkyl-alkylcarbonylamino, cyano, (C₃-C₇)cycloalkyl, (C₃-C₇)heterocyclyl, trialkylsilyl, trialkylsilyloxy, phenyl, substituted phenyl, heteroaryl or substituted heteroaryl, C2-C₂₀alkynyl, C₂-C₂₀alkynyl substituted by halogen, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, alkoxy, thioalkyl, alkylthiocarbonyl, alkylcarbonylthio, alkylsulfonyl, alkylsulfoxyl, alkylaminosulfonyl, dialkylaminosulfonyl, alkylsulfonyloxy, alkylsulfonylamino, alkylamino, dialkylamino, alkylcarbonylamino, dialkylcarbonylamino, alkylalkylcarbonylamino, cyano, (C₃-C₇)cycloalkyl, (C₃-C₇)heterocyclyl, trialkylsilyl, trialkylsilyloxy. phenyl, substituted phenyl, heteroaryl or substituted heteroaryl, (C1-C7)cycloalkyl, (C1-C₇)cycloalkyl substituted by halogen, haloalkyl, (C₁-C₆)alkyl, alkoxy, alkylcarbonyloxy. thioalkyl, alkylcarbonylthio, alkylamino, alkylcarbonylamino, trialkylsilyloxy, heteroaryl, heteroaryl substituted by halogen, haloalkyl, nitro, cyano, (C1-C6)alkyl, alkoxy, alkylcarbonyloxy, thioalkyl, alkylcarbonylthio, alkylamino, alkylcarbonylamino, trialkylsilyl or trialkylsilyloxy, heteroaryloxy, substituted heteroaryloxy, heteroarylthio, substituted heteroarylthio, heteroarylamino, substituted heteroarylamino, diheteroarylamino, substituted diheteroarylamino, phenylamino, substituted phenylamino, diphenylamino, substituted diphenylamino, cycloalkylamino, substituted cycloalkylamino, dicycloalkylamino, substituted dicycloalkylamino, cycloalkoxy or substituted cycloalkoxy, and salts and diastereomers of the compounds of the formula I.

2. A compound according to claim 1, wherein R4 and R5 together are a group

$$-C-R_6(R_7)-O-C-R_8(R_9)-C-R_{10}(R_{11})-C-R_{12}(R_{13})-$$
 (Z₁),

$$-C-R_{14}(R_{15})-C-R_{16}(R_{17})-O-C-R_{18}(R_{19})-C-R_{20}(R_{21})-$$
 (Z₂), or

$$-C-R_{22}(R_{23})-C-R_{24}(R_{25})-C-R_{26}(R_{27})-O-C-R_{28}(R_{29})-$$
 (Z₃);

in which R_6 , R_7 , R_8 , R_9 , R_{10} , R_{11} , R_{12} , R_{13} , R_{14} , R_{15} , R_{16} , R_{17} , R_{18} , R_{19} , R_{20} , R_{21} , R_{22} , R_{23} , R_{24} , R_{25} , R_{26} , R_{27} , R_{28} and R_{29} independently of one another are hydrogen, halogen, C_1 - C_4 alkyl or C_1 - C_4 haloalkyl, where an alkylene ring which, together with the carbon atoms of the groups Z_1 , Z_2 and Z_3 , contains 3 to 6 carbon atoms may be fused or spiro-linked to the groups Z_1 , Z_2 and Z_3 .

- 3. A compound according to claim 1, wherein G is hydrogen.
- 4. A compound according to claim 1, wherein R₄ and R₅ together are a group Z₂.

- 5. A compound according to claim 1, wherein at least one ring atom of the groups Z_1 , Z_2 or Z_3 is bridged by an alkylene ring which, together with the carbon atoms of the groups Z_1 , Z_2 or Z_3 , contains 2 to 6 carbon atoms and may be interrupted by oxygen.
- 6. A compound according to claim 1, wherein R_1 , R_2 and R_3 independently of one another are halogen, C_1 - C_4 alkyl, C_2 - C_4 alkenyl, C_2 - C_4 alkynyl or C_1 - C_6 alkoxy.
- 7. A compound according to claim 1, wherein R₂ is halogen, methyl, ethyl or ethinyl.
- 8. A compound according to claim 1, wherein G is the group $-C(X_1)-R_{30}$ or $C(X_2)-(X_3)-R_{31}$ in which X_1 , X_2 and X_3 are oxygen and R_{30} and R_{31} independently of one another are C_1-C_5 alkyl.
- 9. A compound according to claim 1, wherein R_1 and R_3 independently of one another are methyl, ethyl, isopropyl, vinyl, allyl, ethinyl, methoxy, ethoxy, bromine or chlorine.
- 10. A compound according to claim 1, wherein R_{30} , R_{31} , R_{32} , R_{33} , R_{34} , R_{35} and R_{36} independently of one another are hydrogen, C_1 - C_5 alkyl or C_1 - C_5 haloalkyl.
- 11. A herbicidal and plant-growth-inhibiting composition, which contains a herbicidally effective amount of a compound of the formula I on an inert carrier.
- 12. A method for controlling undesirable plant growth, wherein a herbicidally effective amount of an active compound of the formula I or a composition which contains this active compound is applied to the plants or their habitat.
- 13. A method for inhibiting plant growth, wherein a herbididally effective amount of an active compound of the formula I or a composition which contains this active compound is applied to the plants or their habitat.
- 14. A selective-herbicidal composition which comprises as active compound, in addition to customary inert formulation auxiliaries, a mixture of
- a) a herbicidally effective amount of a compound of the formula I according to claim 1 and
- b) a herbicide-antagonistically effective amount of either a compound of the formula X

$$\begin{array}{c}
X_6 \\
O - CH_2 - O - R_{37}
\end{array}$$
(X),

in which R_{37} is hydrogen, C_1 - C_8 alkyl or C_1 - C_6 alkoxy- or C_3 - C_6 alkenyloxy-substituted C_1 - C_8 alkyl; and X_6 is hydrogen or chlorine; or a compound of the formula XI

in which

E is nitrogen or methine; R_{38} is -CCl₃, phenyl or halogen-substituted phenyl; R_{39} and R_{40} independently of one another are hydrogen or halogen; and R_{41} is C_1 - C_4 alkyl; or a compound of the formula XII

in which R_{44} and R_{45} independently of one another are hydrogen or halogen and R_{46} , R_{47} and R_{48} independently of one another are C_1 - C_4 alkyl, or a compound of the formula XIII

$$R_{51}$$
 N -CO-N
 R_{52}
 R_{53}
 R_{53}
 R_{50}
 R_{50}

in which A2 is a group

R₅₁ and R₅₂ independently of one another are hydrogen, C₁-C₈alkyl, C₃-C₈cycloalkyl,

$$C_3$$
- C_6 alkenyl, C_3 - C_6 alkynyl, R_y , or C_1 - C_4 alkoxy- or R_y

substituted C_1 - C_4 alkyl; or R_{51} and R_{52} together form a C_4 - C_6 alkylene bridge which may be interrupted by oxygen, sulfur, SO, SO₂, NH or -N(C_1 - C_4 alkyl)-,

R₅₃ is hydrogen or C₁-C₄alkyl;

 R_{49} is hydrogen, halogen, cyano, trifluoromethyl, nitro, C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfinyl, -COOR_j, -CONR_kR_m, -COR_n, -SO₂NR_kR_m or -OSO₂- C_1 - C_4 alkyl;

 R_g is hydrogen, halogen, cyano, nitro, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, -COOR_j, -CONR_kR_m, -COR_n, -SO₂NR_kR_m, -OSO₂- C_1 - C_4 alkyl, C_1 - C_6 alkoxy, or C_1 - C_6 alkoxy which is substituted by C_1 - C_4 alkoxy or halogen, C_3 - C_6 alkenyloxy, or C_3 - C_6 alkenyloxy which is substituted by halogen, or C_3 - C_6 alkynyloxy,

or R_{49} and R_{50} together form a C_3 - C_4 alkylene bridge which may be substituted by halogen or C_1 - C_4 alkyl, or they form a C_3 - C_4 alkenylene bridge which may be substituted by halogen or C_1 - C_4 alkyl, or they form a C_4 alkadienylene bridge which may be substituted by halogen or C_1 - C_4 alkyl;

 R_{50} and R_h independently of one another are hydrogen, halogen, C_1 - C_4 alkyl, trifluoromethyl, C_1 - C_6 alkoxy, C_1 - C_6 alkylthio or -COOR_i;

 R_c is hydrogen, halogen, nitro, C_1 - C_4 alkyl or methoxy; R_d is hydrogen, halogen, nitro, C_1 - C_4 alkyl, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, -COOR $_i$ or CONR $_k$ R $_m$;

 R_e is hydrogen, halogen, C_1 - C_4 alkyl, -COOR $_j$, trifluoromethyl or methoxy, or R_d and R_e together form a C_3 - C_4 alkylene bridge;

Rp is hydrogen, halogen, C₁-C₄alkyl, -COOR_j, trifluoromethyl or methoxy; Rq is hydrogen, halogen, nitro, C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, -COOR_j or CONR_kR_m, or Rp and Rq together form a C₃-C₄alkylene bridge;

Rr is hydrogen, halogen, C₁-C₄alkyl, -COOR_j, trifluoromethyl or methoxy; Rs is hydrogen, halogen, nitro, C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, -COOR_j or CONR_kR_m, or Rr and Rs together form a C₃-C₄alkylene bridge;

Rt is hydrogen, halogen, C₁-C₄alkyl, -COOR_j, trifluoromethyl or methoxy; Ru is hydrogen, halogen, nitro, C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, -COOR_j or CONR_kR_m, or Rv and Ru together form a C₃-C₄alkylene bridge;

R₁ and Rv are hydrogen, halogen or C₁-C₄alkyl;

 R_x and R_y independently of one another are hydrogen, halogen, C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylthio, -COOR₅₄, trifluoromethyl, nitro or cyano;

 R_{j} , R_{k} and R_{m} independently of one another are hydrogen or C_{1} - C_{4} alkyl; or R_{k} and R_{m} together form a C_{4} - C_{6} alkylene bridge which may be interrupted by oxygen, NH or -N(C_{1} - C_{4} alkyl)-;

 R_n is C_1 - C_4 alkyl, phenyl, or halogen-, C_1 - C_4 alkyl-, methoxy-, nitro- or trifluoromethyl-substituted phenyl;

 R_{54} is hydrogen, C_1 - C_{10} alkyl, C_1 - C_4 alkoxy- C_1 - C_4 alkyl, C_1 - C_4 alkylthio- C_1 - C_4 alkyl, di- C_1 - C_4 alkylamino- C_1 - C_4 alkyl, halo- C_1 - C_8 alkyl, C_2 - C_8 alkenyl, halo- C_2 - C_8 alkenyl, C_3 - C_7 cycloalkyl, halo- C_3 - C_7 cycloalkyl, C_1 - C_8 alkylcarbonyl, allylcarbonyl, C_3 - C_7 cycloalkylcarbonyl, benzoyl which is unsubstituted or substituted up to three times on the

phenyl ring by identical or different substituents selected from the group consisting of

halogen, C_1 - C_4 alkyl, halo- C_1 - C_4 alkyl, halo- C_1 - C_4 alkyl, halo- C_1 - C_4 alkoxy or C_1 - C_4 alkoxy; or furoyl, thienyl; or C_1 - C_4 alkyl which is substituted by phenyl, halophenyl, C_1 - C_4 alkylphenyl, C_1 - C_4 alkoxyphenyl, halo- C_1 - C_4 alkylphenyl, halo- C_1 - C_4 alkoxyphenyl, C_1 - C_6 alkoxycarbonyl, C_1 - C_6 alkoxycarbonyl, C_3 - C_8 alkenyloxycarbonyl, C_3 - C_8 alkylphiocarbonyl, C_3 - C_8 alkenylthiocarbonyl, C_3 - C_8 alkylphiocarbonyl, carbamoyl, mono- C_1 - C_4 alkylaminocarbonyl, di- C_1 - C_4 alkylaminocarbonyl; or phenylaminocarbonyl which is unsubstituted or substituted up to three times on the phenyl by identical or different substituents selected from the group consisting of halogen, C_1 - C_4 alkyl, halo- C_1 - C_4 alkyl, halo- C_1 - C_4 alkoxy and C_1 - C_4 alkoxy, or is monosubstituted by cyano or nitro, or dioxolan-2-yl which is unsubstituted or substituted by one or two C_1 - C_4 alkyl radicals, or dioxan-2-yl which is unsubstituted or substituted by one or two C_1 - C_4 alkyl radicals, or C_1 - C_4 alkyl which is substituted by cyano, nitro, carboxyl or C_1 - C_8 alkylthio- C_1 - C_8 alkoxycarbonyl; or a compound of the formula XIV

$$R_{56}$$
 N $CHCl_2$ (XIV),

in which R₅₆ and R₅₇ independently of one another are C₁-C₆alkyl or C₂-C₆alkenyl; or R₅₆ and

$$R_{58}$$
 , R_{59} ; R_{58} and R_{59} independently of one another are hydrogen

or
$$C_1$$
- C_6 alkyl; or R_{56} and R_{57} together are

 R_{60} and R_{61} independently of one another are C_1 - C_4 alkyl, or R_{60} and R_{61} together are -(CH_2)₅-;

R₆₂ is hydrogen, C₁-C₄alkyl or

or R_{56} and R_{57} together are

or
$$R_{72}$$
 R_{74}
 R_{75}
 R_{76}
 R_{77}
 R_{76}

 R_{63} , R_{64} , R_{65} , R_{66} , R_{67} , R_{68} , R_{69} , R_{70} , R_{71} , R_{72} , R_{73} , R_{74} , R_{75} , R_{76} , R_{77} and R_{78} independently of one another are hydrogen or C_1 - C_4 alkyl; or a compound of the formula XV

$$R_{80}$$
 $N-O$
 O
 O
 (XV)

in which R_{80} is hydrogen or chlorine and R_{79} is cyano or trifluoromethyl, or a compound of the formula XVI

$$R_{81} \longrightarrow N \longrightarrow CI \qquad (XVI)$$

in which R_{81} is hydrogen or methyl, or of the formula XVII

$$R_{83}$$

$$R_{84}$$

$$Z_{4}$$

$$W_{1}$$

$$(XVII),$$

in which

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 R_{82} is hydrogen, C_1 - C_4 alkyl, C_1 - C_4 alkyl which is substituted by C_1 - C_4 alkyl- X_2 - or C_1 - C_4 haloalkyl- X_2 -, C_1 - C_4 haloalkyl, nitro, cyano, -COOR₈₅, -NR₈₆R₈₇, -SO₂NR₈₈R₈₉ or -CONR₉₀R₉₁;

 R_{83} is hydrogen, halogen, C_1 - C_4 alkyl, trifluoromethyl, C_1 - C_4 alkoxy or C_1 - C_4 haloalkoxy; R_{84} is hydrogen, halogen or C_1 - C_4 alkyl;

U, V, W_1 and Z_4 independently of one another are oxygen, sulfur, $C(R_{92})R_{93}$, carbonyl, NR_{94} ,

a group
$$C = \begin{matrix} O \\ A_1 \end{matrix}$$
 or $C = \begin{matrix} C \\ C \end{matrix}$ R_{102} in which R_{102} is C_2 - C_4 alkenyl or R_{95}

C₂-C₄alkynyl; with the proviso that

a) at least one of the ring members U, V, W_1 or Z_4 is carbonyl, and a ring member which is

adjacent to this or these ring members is the group

$$C = C$$
 R_{102}
 R_{96} , this group being present only once; and

b) two adjacent ring members U and V, V and W_1 and W_1 and Z_4 may not simultaneously be oxygen;

 R_{95} and R_{96} independently of one another are hydrogen or C_1 - C_8 alkyl; or R_{95} and R_{96} together form a C_2 - C_6 alkylene group;

 A_1 is R_{99} - Y_1 - or -NR₉₇ R_{98} ;

X₂ is oxygen or -S(O)_s;

Y₁ is oxygen or sulfur;

 R_{99} is hydrogen, C_1 - C_8 alkyl, C_1 - C_8 haloalkyl, C_1 - C_4 alkoxy- C_1 - C_8 alkyl, C_3 - C_6 alkenyloxy- C_1 - C_8 alkyl or phenyl- C_1 - C_8 alkyl, where the phenyl ring may be substituted by halogen, C_1 - C_4 alkyl, trifluoromethyl, methoxy or methyl- $S(O)_5$ -, C_3 - C_6 alkenyl, C_3 - C_6 haloalkenyl, phenyl- C_3 - C_6 alkynyl, phenyl- C_3 - C_6 alkynyl, oxetanyl, furyl or tetrahydrofuryl; R_{85} is hydrogen or C_1 - C_4 alkyl;

R₈₆ is hydrogen, C₁-C₄alkyl or C₁-C₄alkylcarbonyl;

R₈₇ is hydrogen or C₁-C₄alkyl; or

R₈₆ and R₈₇ together form a C₄- or C₅alkylene group;

 R_{88} , R_{89} , R_{90} and R_{91} independently of one another are hydrogen or C_1 - C_4 alkyl; or R_{88} together with R_{89} or R_{90} together with R_{91} independently of one another are C_4 - or C_5 -alkylene, where a carbon atom may be replaced by oxygen or sulfur, or one or two carbon atoms may be replaced by -NR₁₀₀-;

 R_{92} , R_{100} and R_{93} independently of one another are hydrogen or C_1 - C_8 alkyl; or R_{92} and R_{93} together are C_2 - C_6 alkylene;

R₉₄ is hydrogen or C₁-C₈alkyl;

 R_{97} is hydrogen, C_1 - C_8 alkyl, phenyl, phenyl- C_1 - C_8 alkyl, where the phenyl rings may be substituted by fluorine, chlorine, bromine, nitro, cyano, -OCH₃, C_1 - C_4 alkyl or CH_3SO_2 -, C_1 - C_4 alkoxy- C_1 - C_8 alkyl, C_3 - C_6 alkenyl or C_3 - C_6 alkynyl;

R₉₈ is hydrogen, C₁-C₈alkyl, C₃-C₆alkenyl or C₃-C₆alkynyl; or

R₉₇ and R₉₈ together are C₄- or C₅-alkylene, where a carbon atom may be replaced by oxygen or sulfur, or one or two carbon atoms may be replaced by -NR₁₀₁-;

R₁₀₁ is hydrogen or C₁-C₄alkyl;

r is 0 or 1; and

s is 0, 1 or 2, or a compound of the formula XVIII

in which R_{103} is hydrogen, C_1 - C_6 alkyl, C_3 - C_6 cycloalkyl, C_3 - C_6 alkenyl or C_3 - C_6 alkynyl; and R_{104} , R_{105} and R_{106} independently of one another are hydrogen, C_1 - C_6 alkyl, C_3 - C_6 cycloalkyl or C_1 - C_6 alkoxy, with the proviso that one of the substituents R_{104} , R_{105} and R_{106} is different from hydrogen;

a compound of the formula XIX

in which Z_5 is N or CH, n, in the case where Z_5 is N, is 0, 1, 2 or 3 and, in the case where Z_5 is CH, is 0, 1, 2, 3 or 4, R_{107} is halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkoxy, nitro, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl, C_1 - C_4 alkoxycarbonyl or unsubstituted or substituted phenyl or phenoxy, R_{108} is hydrogen or C_1 - C_4 alkyl, R_{109} is hydrogen, C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, C_2 - C_6 alkenyl, C_2 - C_6 alkenyl, C_1 - C_4 alkyl, C_1 - C_4 alkyl, C_1 - C_4 alkylthio- C_1 - C_4 alkyl, C_1 - C_4 alkylsulfonyl- C_1 - C_4 alkyl, C_1 - C_4 a

$$\mathbf{z}_{\mathbf{0}}$$
 $\mathbf{z}_{\mathbf{0}}$ $\mathbf{z}_{\mathbf{0}}$ $\mathbf{z}_{\mathbf{0}}$

in which Z_6 is O or N-R₁₁₀ and R₁₁₀ is a group of the formula

in which R_{111} and R_{112} independently of one another are cyano, hydrogen, C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, C_2 - C_6 alkenyl, unsubstituted or substituted phenyl or heteroaryl;

a compound of the formula XXI
$$Z_7$$
 R_{114} R_{113} (XXI),

in which Z_7 is O, S, S=O, SO₂ or CH₂, R₁₁₃ and R₁₁₄ independently of one another are hydrogen, halogen or C₁-C₄alkyl, W₂ and W₃ independently of one another are CH₂COOR₁₁₅, COOR₁₁₅ or together are a group of the formula -(CH₂)C(O)-O-C(O)-(CH₂)-, and R₁₁₅ is hydrogen, C₁-C₄alkyl, C₂-C₄alkenyl, C₂-C₆alkynyl, C₃-C₆cycloalkyl, C₁-C₄haloalkyl, a metal cation or an ammonium cation; a compound of the formula XXII

in which R_{119} and R_{120} independently of one another are hydrogen, halogen or C_1 - C_4 haloalkyl, R_{121} is hydrogen, C_1 - C_4 alkyl, C_3 - C_4 alkenyl, C_3 - C_4 alkynyl, C_1 - C_4 haloalkyl, C_3 - C_6 cycloalkyl, a metal cation or an ammonium cation, Z_8 is N, CH, C-F or C-Cl and W_4 is a group of the formula

in which R_{122} and R_{123} independently of one another are hydrogen or C_1 - C_4 alkyl and R_{124} and R_{125} independently of one another are hydrogen or C_1 - C_4 alkyl; a compound of the formula XXIII

in which R_{126} is hydrogen, cyano, halogen, C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkoxycarbonyl, C_1 - C_4 alkylthiocarbonyl, -NH- R_{128} , -C(O)NH- R_{128} , unsubstituted or substituted aryl or heteroaryl,

 R_{127} is hydrogen, cyano, nitro, halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 thioalkyl, C_1 - C_4 haloalkyl, -NH- R_{128} , -C(O)NH- R_{128} , unsubstituted or substituted aryl,

heteroaryl, and R_{128} is C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_3 - C_4 alkenyl, C_3 - C_4 alkynyl, C_3 - C_4 cycloalkyl, unsubstituted or substituted aryl or heteroaryl, formyl, C_1 - C_4 -alkylcarbonyl, C_1 - C_4 -alkylsulfonyl;

a compound of the formula XXIV

$$R_{132}$$
 R_{133}
 R_{130}
 R_{130}
 R_{129}
(XXIV),

in which R_{129} and R_{130} independently of one another are hydrogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, mono- C_1 - C_8 - or di- C_1 - C_8 alkylamino, C_3 - C_6 cycloalkyl, C_1 - C_4 thioalkyl, phenyl or heteroaryl, R_{131} has the meaning of R_{129} and is additionally OH, NH₂, halogen, di- C_1 - C_4 aminoalkyl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl or C_1 - C_4 alkoxycarbonyl, R_{132} has the meaning of R_{129} and is additionally cyano, nitro, carboxyl, C_1 - C_4 alkoxycarbonyl, di- C_1 - C_4 aminoalkyl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl, SO_2 -OH,

iso- C_1 - C_4 aminoalkylsulfonyl or C_1 - C_4 alkoxysulfonyl, R_{133} has the meaning of R_{129} and is additionally OH, NH₂, halogen, di- C_1 - C_4 aminoalkyl, pyrrolidin-1-yl, piperidin-1-yl, morpholin-1-yl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl, C_1 - C_4 alkoxycarbonyl, phenoxy, naphtoxy, phenylamino, benzoyloxy or phenylsulfonyloxy;

or a compound of the formula XXV

$$R_{136}$$
 O R_{134} R_{135} (XXV) ,

in which R_{134} is hydrogen, C_4 alkyl, C_1 - C_4 haloalkyl, C_2 - C_4 alkenyl, C_2 - C_4 alkynyl or C_1 - C_4 alkoxy- C_1 - C_4 alkyl, R_{135} is hydrogen, halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or C_1 - C_4 alkoxy and R_{136} is hydrogen, halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or C_1 - C_4 alkoxy, with the proviso that R_{135} and R_{136} are not simultaneously hydrogen.

15. A composition according to claim 14, wherein it comprises, as herbicide-antagonistically effective amount, either a compound of the formula X

$$\begin{array}{c}
X_6 \\
O - CH_2 - O - R_{37}
\end{array}$$
(X),

in which R_{37} is hydrogen, C_1 - C_8 alkyl or C_1 - C_6 alkoxy- or C_3 - C_6 alkenyloxy-substituted C_1 - C_8 alkyl; and X_6 is hydrogen or chlorine; or a compound of the formula XI

in which

E is nitrogen or methine; R_{38} is -CCl₃, phenyl or halogen-substituted phenyl; R_{39} and R_{40} independently of one another are hydrogen or halogen; and R_{41} is C_1 - C_4 alkyl; or a compound of the formula XII

$$R_{47}O_2C$$
 N
 R_{46}
 N
 R_{45}
 R_{44}
 N
 N
 N
 N

in which R_{44} and R_{45} independently of one another are hydrogen or halogen and R_{46} , R_{47} and R_{48} independently of one another are C_1 - C_4 alkyl.

16. A method for the selective control of weeds and grasses in crops of useful plants, wherein the useful plants, their seeds or seedlings or the area on which they are cultivated are treated with a herbicidally effective amount of a herbicide of the formula I and a

- 17. A composition according to claim 11, which contains spray tank adjuvants.
- 18. A composition according to claim 14, which contains spray tank adjuvants.

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A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C07D498/04 A01N43/56 A01N43/90 According to international Patent Classification (IPC) or to both national classification and IPC B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) IPC 7 C07D Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practical, search terms used) CHEM ABS Data C. DOCUMENTS CONSIDERED TO BE RELEVANT Relevant to claim No. Citation of document, with indication, where appropriate, of the relevant passages Category 3 1,11,14 WO 96 25395 A (BAYER AKTIENGESELLSCHAFT) Υ 22 August 1996 (1996-08-22) cited in the application page 1 -page 15, line 7 WO 96 21652 A (CIBA-GEIGY AG) 1,11,14 Υ 18 July 1996 (1996-07-18) cited in the application page 1 -page 8 1,11,14 WO 96 11574 A (CIBA-GEIGY) Υ 25 April 1996 (1996-04-25) the whole document 1.11.14 EP 0 508 126 A (BAYER AG) Υ 14 October 1992 (1992-10-14) cited in the application the whole document -/--Patent family members are listed in annex. Х X Further documents are listed in the continuation of box C. or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention Special categories of cited documents: "T" later document published after the international filing date "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the citation or other special reason (as specified) document is combined with one or more other such docu-ments, such combination being obvious to a person skilled "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filling date but "&" document member of the same patent family later than the priority date claimed Date of mailing of the international search report Date of the actual completion of the international search 1 8. JULI 2000 18 July 2000 Authorized officer Name and making address of the ISA European Patent Office, P.B. 5818 Patentiaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040. Tx. 31 651 epo nl. Fax: (+31-70) 340-3016 Kyriakakou, G

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